

Intercontinental Transport of Ozone and Precursors (ITOP)

Ally Lewis, Ruth Purvis, Jim Hopkins, James Lee, Nicola Watson

Department of Chemistry, University of York

Mike Pilling, Dwayne Heard, Trevor Ingham, Lisa Whalley, Cedric Floquet,

Department of Chemistry, University of Leeds

Steve Arnold, Mat Evans, Jim McQuaid, Dominick Spracklen, Kirsty Pringle

School of the Environment, University of Leeds

Paul Monks, Mark Jacob, Alex Parker

Department of Chemistry, University of Leicester

Stuart Penkett, Claire Reeves, Graham Mills, David Oram, Brian Bandy, Debbie Wylding, Jana Slemr,
Dave Stewart, Anne Hulse

School of Environmental Sciences, University of East Anglia

Hugh Coe, Paul Williams, Jonny Crozier, Rami Alfarra

Department of Physics, UMIST

John Methven

Department of Meteorology, University of Reading

John Pyle, Rod Jones, Will Flynn, James Levine, Peter Cook

Department of Chemistry, University of Cambridge

John Reid, Ken Dewey, Nick Price, Steve Devereux, Doug Anderson

Facility for Airborne Atmospheric Measurements

Alan Foster, Alan Roberts, Gaynor Ottoway, Peter Chappell

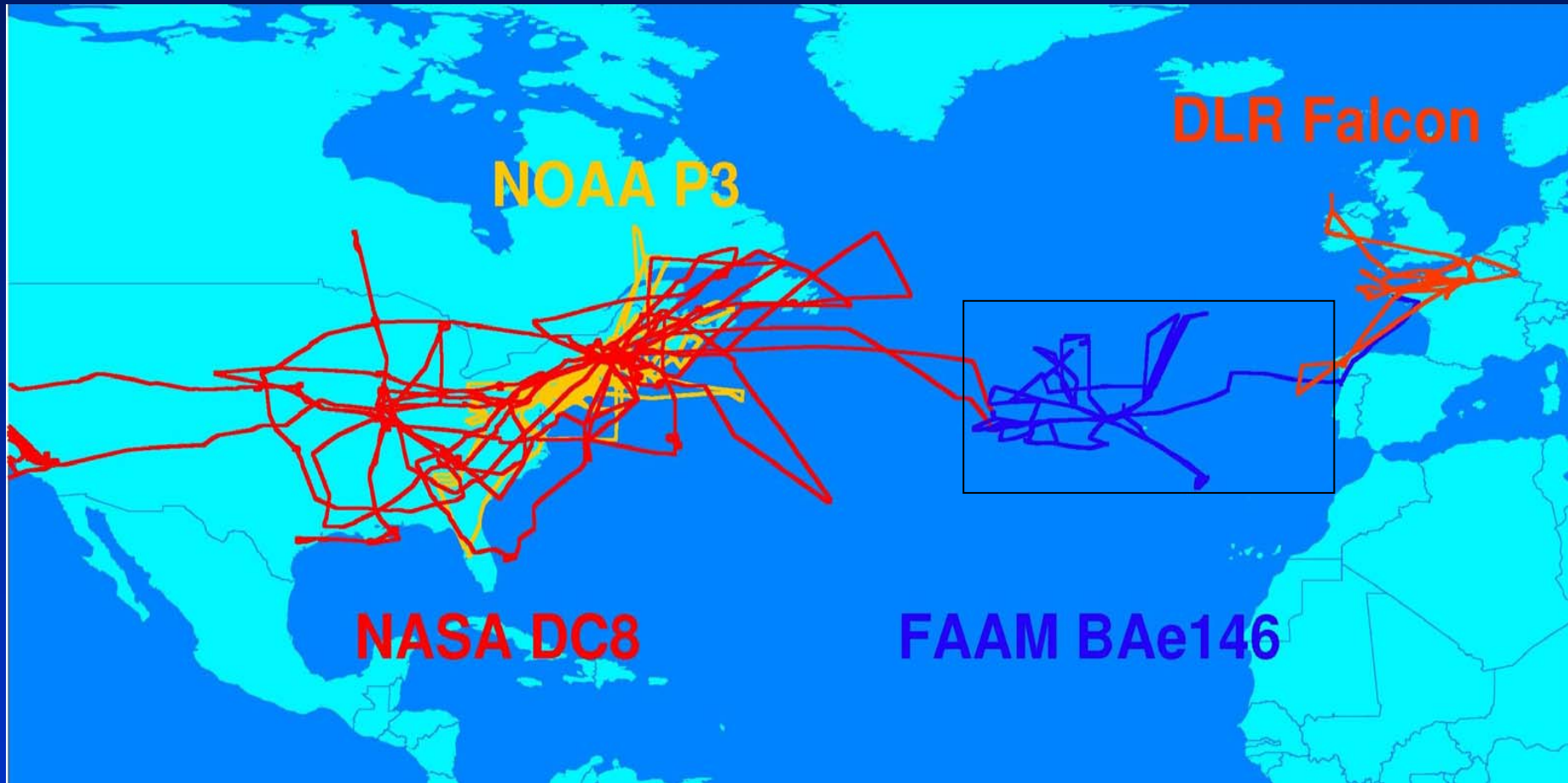
DirectFlight Ltd

Martin Darling, Andrew 'Rodders' Boardman, Simon Tooley

Avalon Aero Ltd

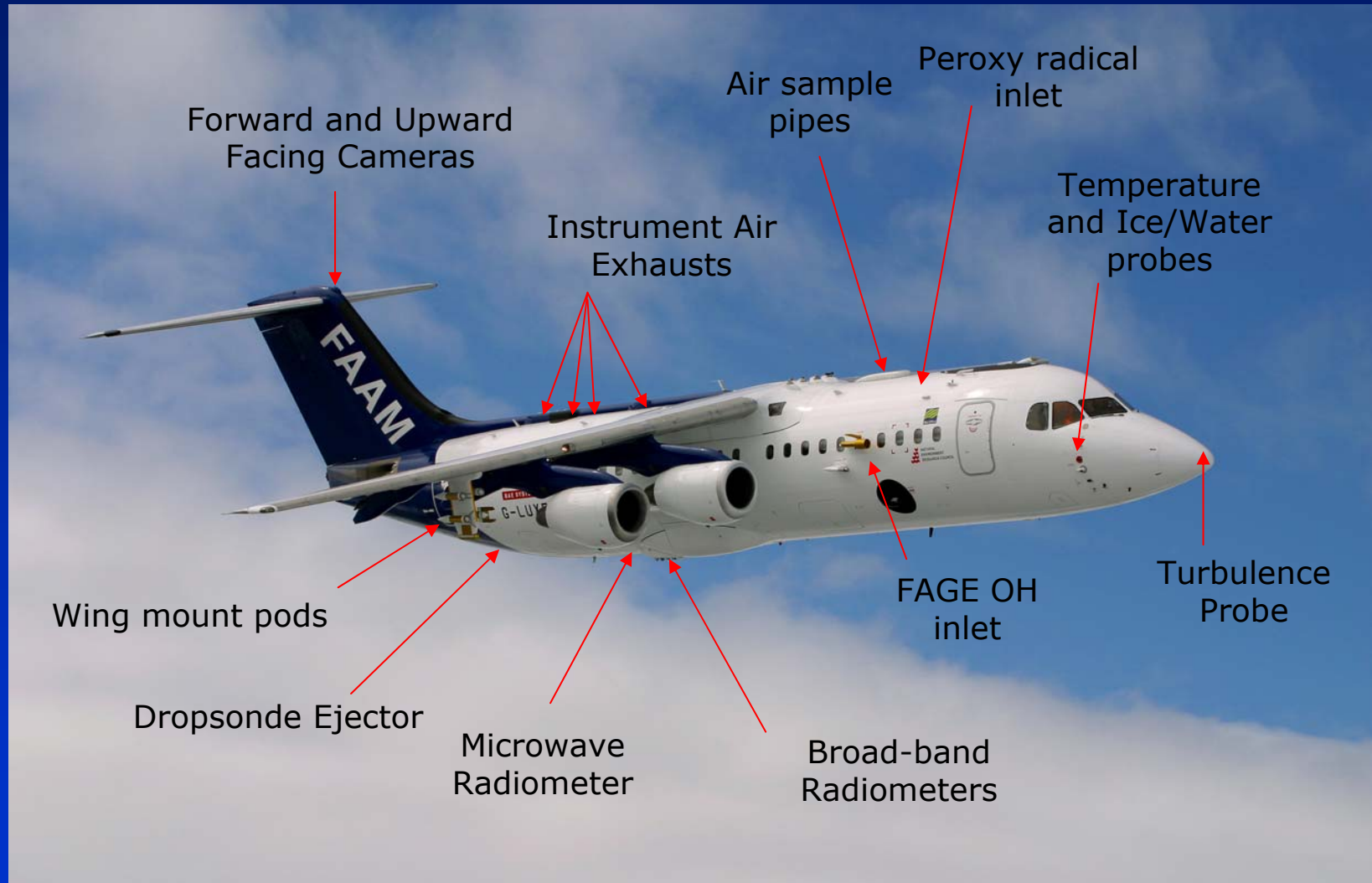


ICARRT Summer 2004



FAAM BAe146, based in Faial, Azores, flew 12 science flights ~70 hours including intercomparison flights with DLR Falcon and NASA DC8

The Facility for Airborne Atmospheric Measurements BAe-146 Research Aircraft



Flight Highlights

- Three-point Lagrangian opportunities: 19/7 + 25/7
28/7 + 29/7 and 31/7 + 1/8.
- Two-point Lagrangian opportunities: 17/7, 20/7 (fire).
- Forest fires: 15/7, 19/7, 20/7, 31/7 (with strat), 1/8.
- Pico flypass: 15/7, 17/7, 19/7, 31/7, 1/8.
- Comparisons: 28/7 (DC8), 3/8 (Falcon).
- ENVISAT underpass: 22/7.

B028 – 12/7/04 – Fire plumes in SW approaches

B029 – 15/7/04 – W to E Low level pollution + forest fires

B030 – 17/7/04 – First Lagrangian opportunity, - skimmed P3 air

B031 – 19/7/04 – New York plume + fire layers

B032 – 20/7/04 – Dominated by Alaskan fires, low T

B033 – 22/7/04 – ENVISAT underpass / cyclone

B034 – 25/7/04 – Re interception of NY plume + African outflow

B035 – 28/7/04 – DC8 comparison / air exported by wcb

B036 – 29/7/04 – Upper level export in wcb from US + fires higher T

B037 – 31/7/04 – low level wcb sampled by P3, + fires + strat

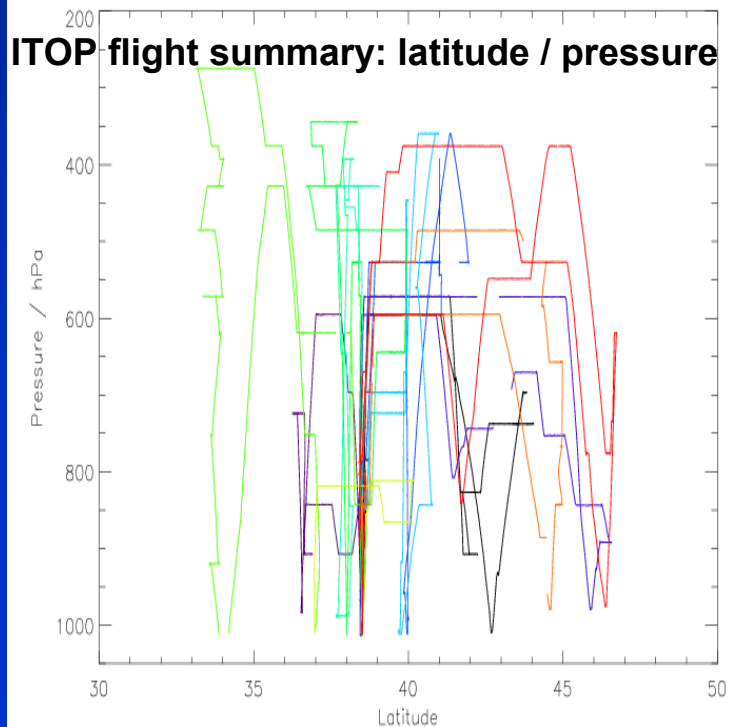
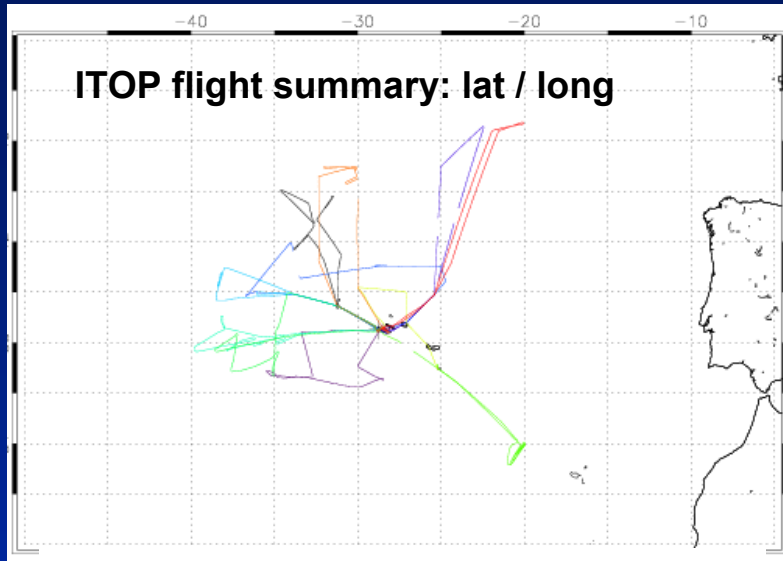
B038 – 01/8/04 – low level wcb 2

B039 – 03/8/04 – Transit / Falcon intercomparison

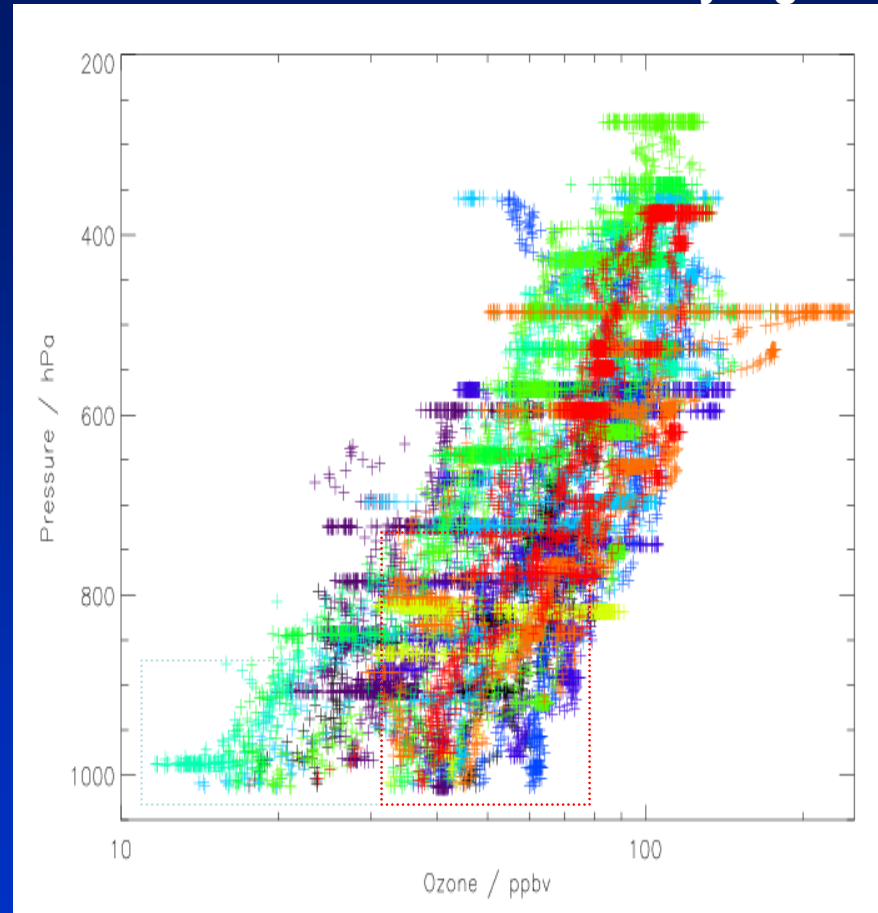
ITOP data coverage

	B028	B029	B030	B031	B032	B033	B034	B035	B036	B037	B038	B039
Core data	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y
CO	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y
O ₃	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y
NO	N	N	N	Y	Y	Y	Y	Y	Y	Y	Y	Y
NO ₂	N	N	N	?	?	?	?	?	?	?	?	?
NO _y	?	?	?	?	?	?	?	?	?	?	?	?
HCHO	N	Y	Y	Y	Y	Y	Y	Y	Y	Y	N	N
H ₂ O ₂	N	N	Y	Y	N	Y	Y	N	Y	Y	N	N
PTR-MS	N	N	N	N	N	N	N	Y	Y	N	N	N
alkyl nitrates	N	N	N	Y	Y	Y	Y	Y	Y	Y	Y	Y
Halocarbons	N	N	N	N	N	N	N	N	N	N	N	Y
PAN	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y
WAS VOCs	26	60	58	61	48	52	59	32	48	61	62	34
OH/HO ₂	N	N	N	N	N	N	N	N	N	N	N	N
radiometers	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y
PERCA	N	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y
AMS	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y
CPC	N	N	Y	Y	Y	Y	N	Y	Y	Y	Y	Y

Overview of U.K. activities



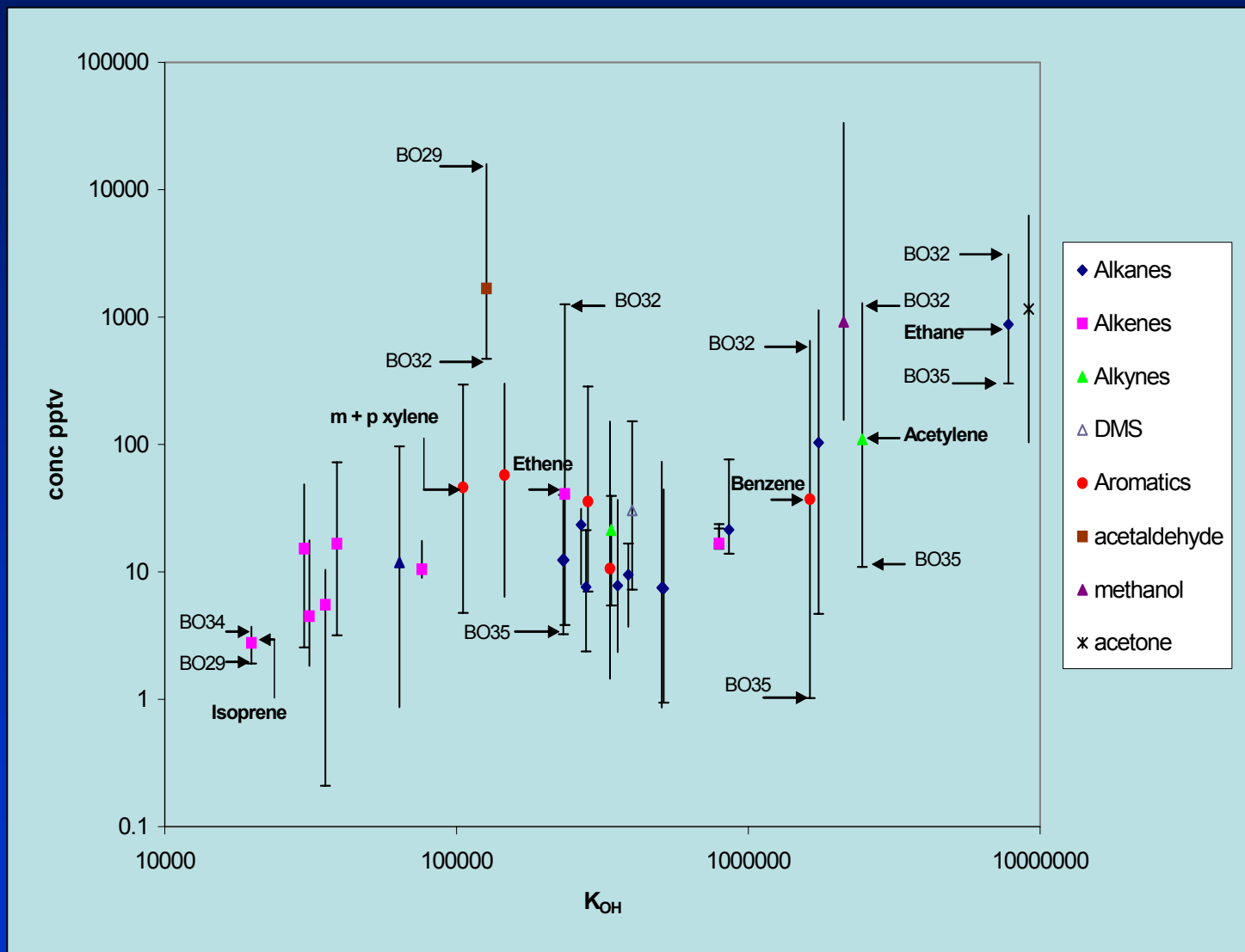
Complete ITOP campaign ozone as a function of altitude. Coloured by flight.



Clean MBL / 15 ppb

Significant long range continental influence at both low and high altitudes during campaign

NMHC variability – campaign overview

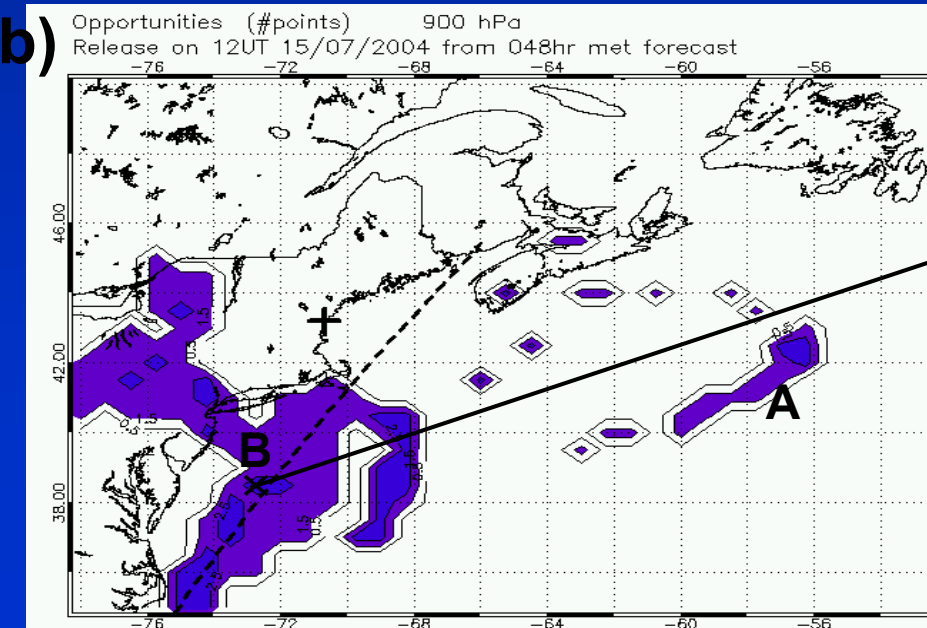
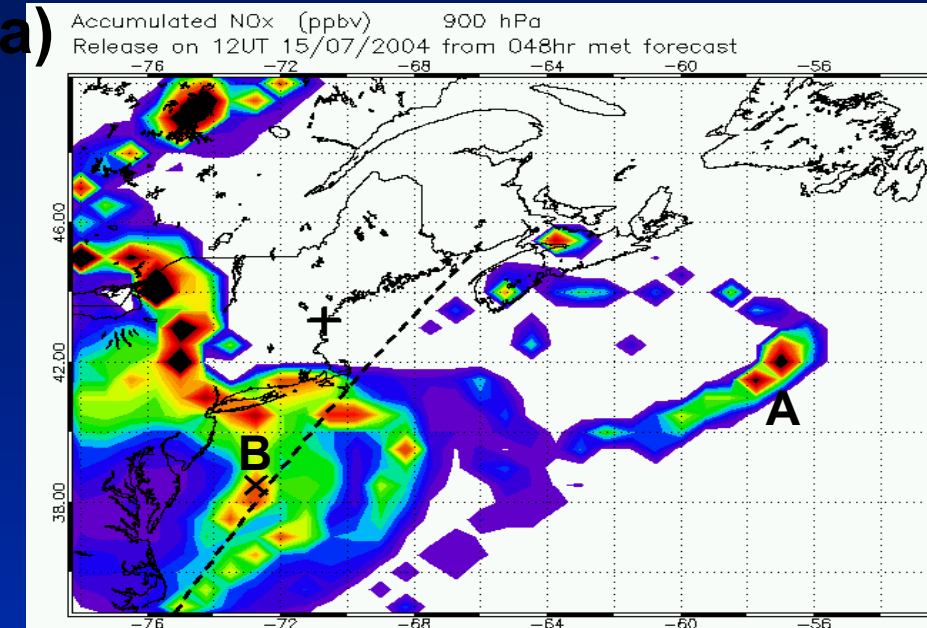


BO29 / 32 fire dominated, BO35 DC8 comparison

Forecasting target for upstream domain

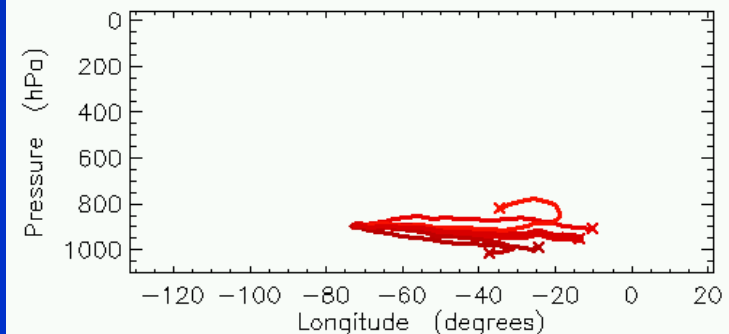
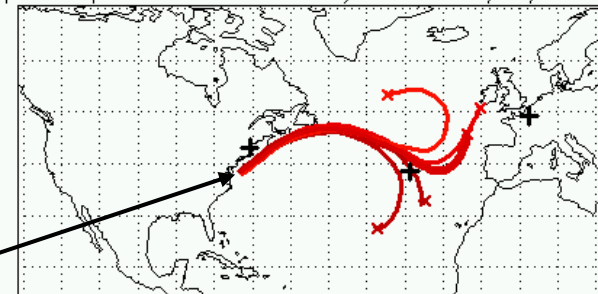
Domain filling trajectory forecasts for E.Coast America domain on 15 July.

- a) NOx emissions tracer at 900 hPa
- b) 2-point and 3-point opportunities highlight targets A and B.
- c) 7 days forward from target B

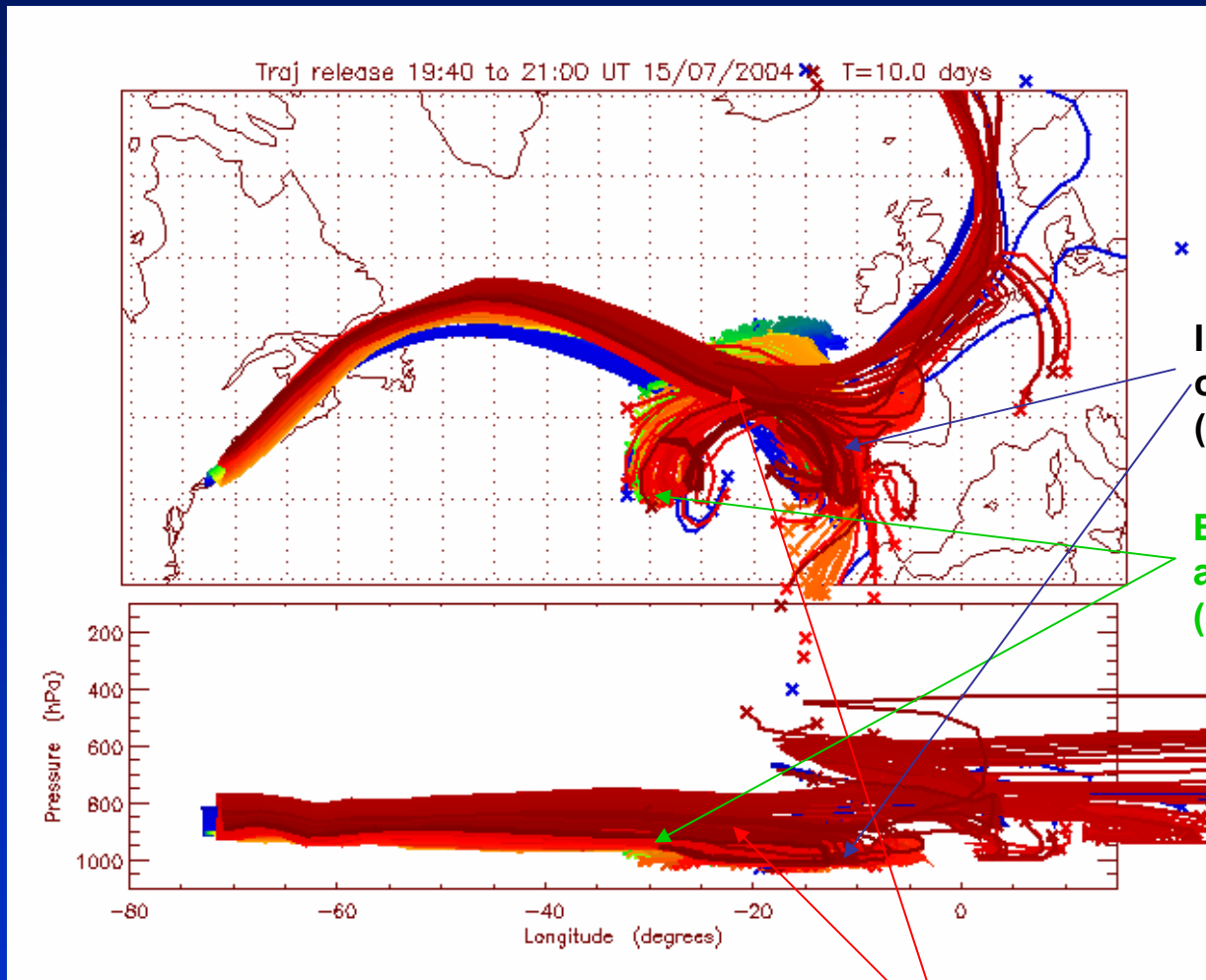


$\lambda=-72$, $\phi=38.5$, $p=900\text{hPa}$, $T=7.00$ days, 12UT 15/07/2004, F+048hr

c)



Forward Trajectories from NOAA P3 flight track



Intercepted by DLR Falcon
off NW corner of Spain on 22/7
(saw CO ~ 90-100ppb).

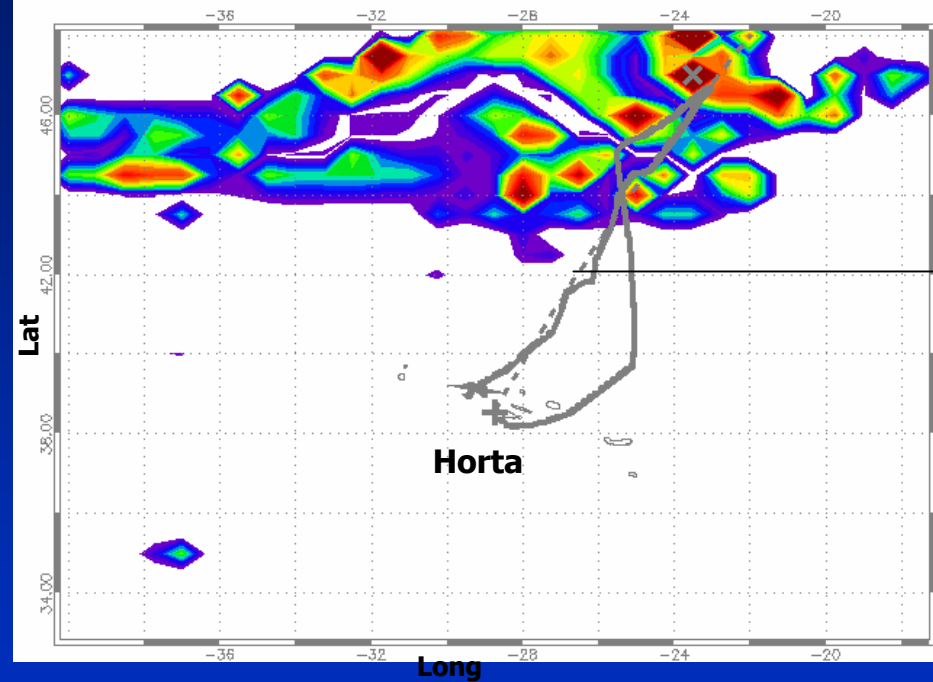
BAE146 flight intercepted this air
again on 25/7
(saw increased CO ~ 115-125ppb).

10 day forward trajectories from NOAA P3
flight off NYC on 15/7. Air mass doubles
back from Spain to the Azores.

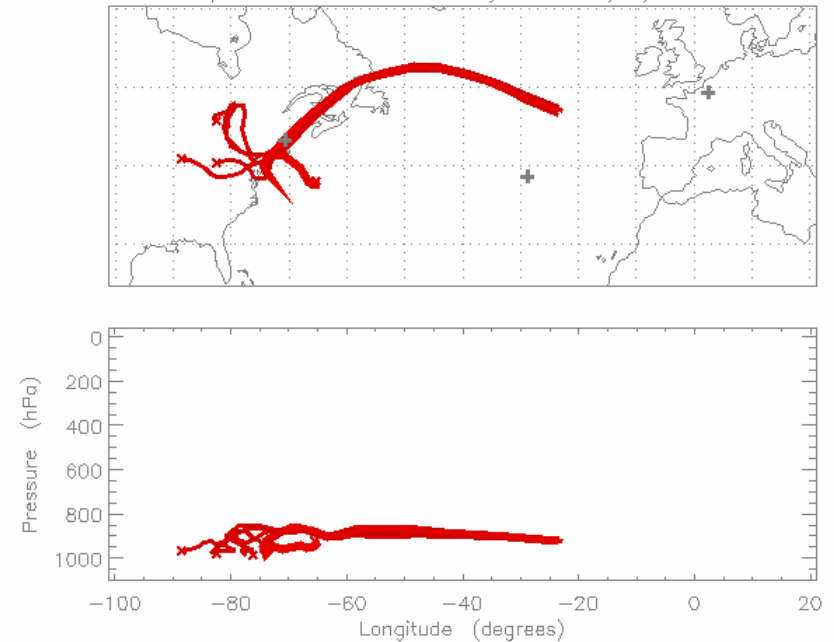
BAE146 flight intercepted this air on 19/7
(saw CO ~ 115-125ppb).

Analysis: Second interception of polluted air mass from USA

Accumulated NO_x (ppbv) 925 hPa
Release on 12UT 19/07/2004 from 000hr met forecast



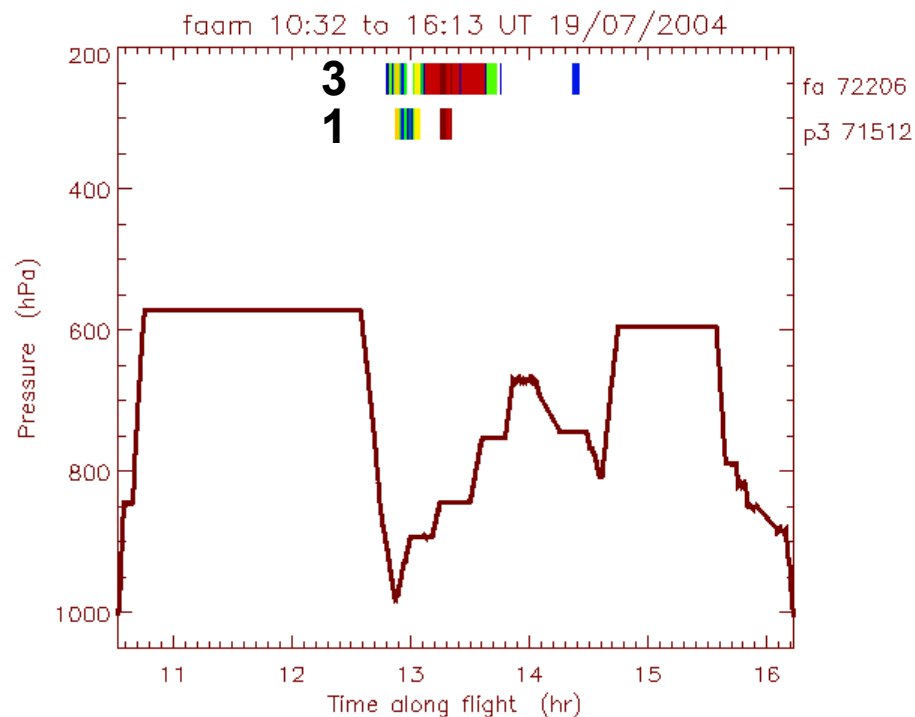
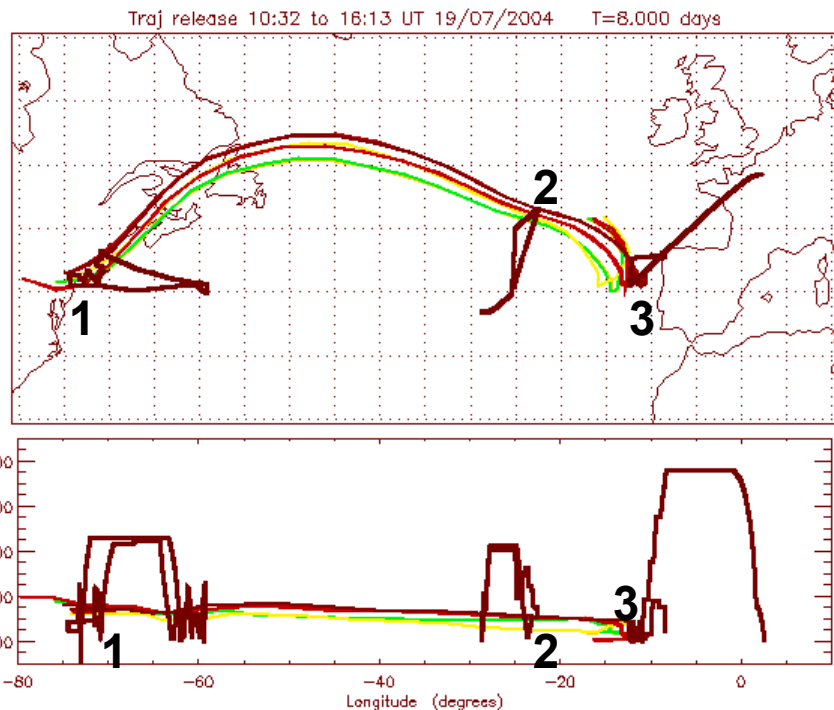
Accumulated NO_x (ppbv) 12UT 19/07/2004 F+000hr
 $\lambda = -23$, $\phi = 47.0$ FL24, T=-7.00 days 12UT 19/07/2004 F+000hr



- NO_x emissions tracer on 925hPa at 12UT 19/7 with BAE146 (air mass relative) flight track overlaid.
- Indicates high emissions into low level outflow from East Coast USA.

Back trajectories from target X confirm NYC as being air mass origin

Did the aircraft make the interception?



Trajectories from BAe146 flight track back and forwards for 4 days.

Best matches with trajectories from other flight tracks.

Time series along flight track.

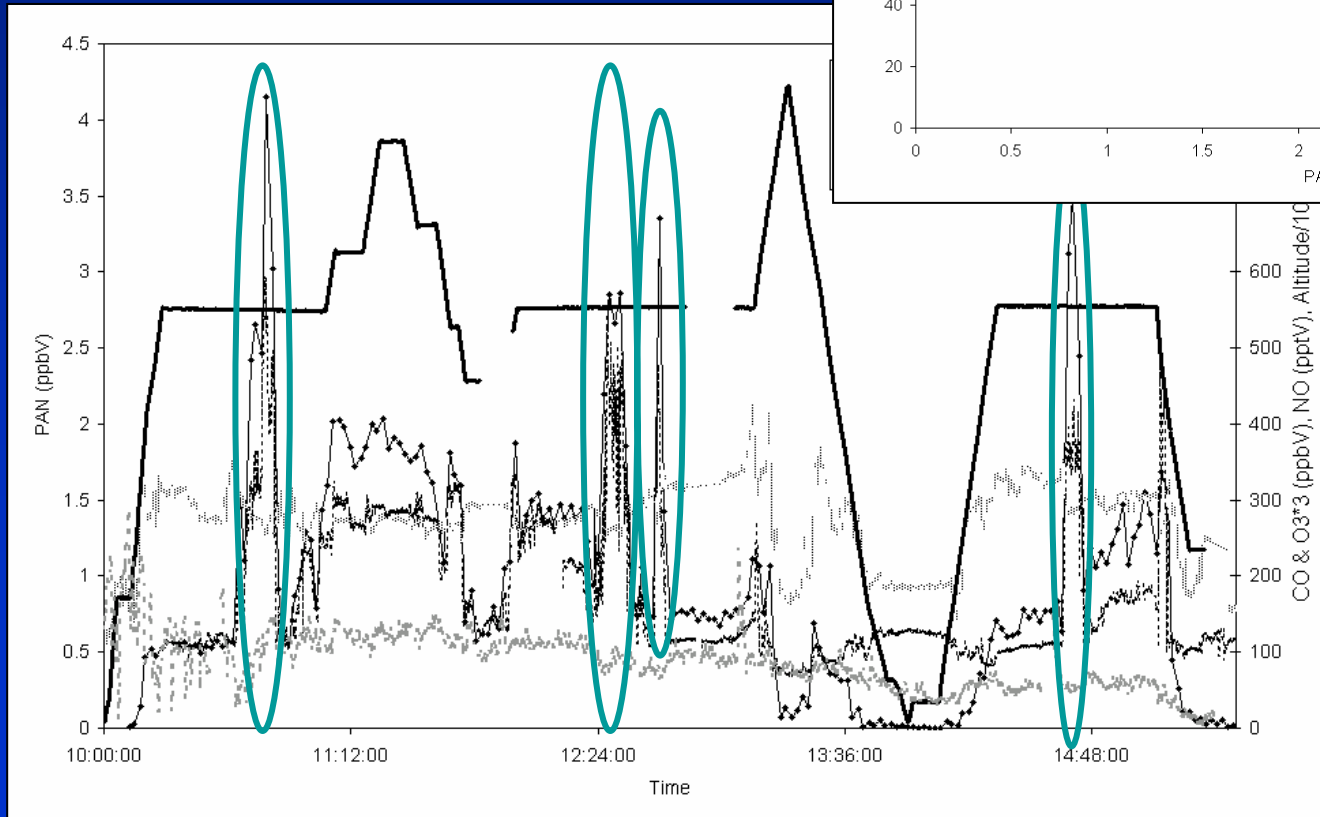
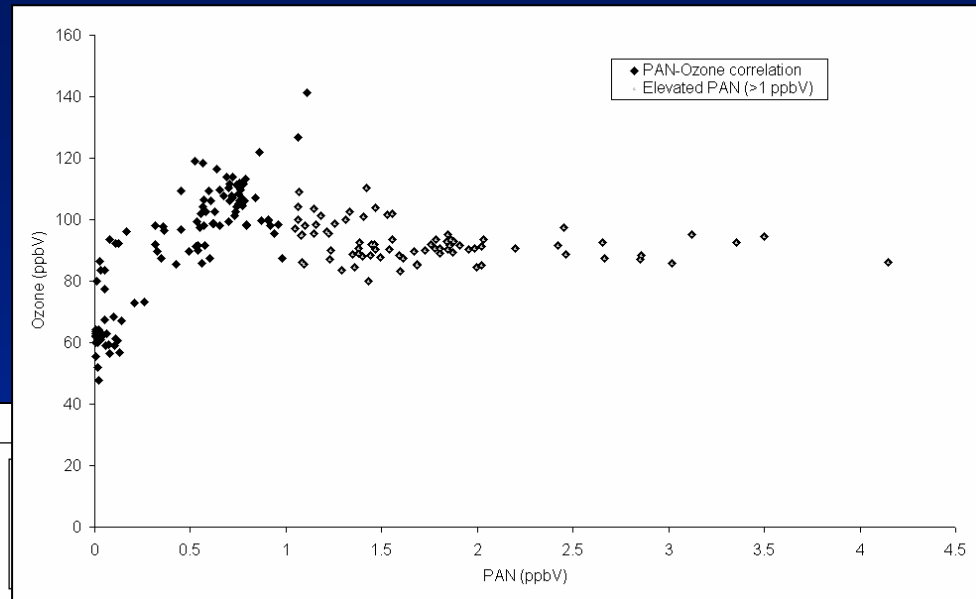
Line shows pressure and colour bars mark air masses intercepted on other flights.

Was the Quasi-Lagrangian expt successful?

- Trajectories based on analyses show that downstream aircraft flew very close to air masses already sampled upstream.
But, can it be regarded as the same air mass?
- 1. Do hydrocarbon fingerprints agree (*using ratios to acetylene to allow for mixing and dilution with surrounding air*)?
- 2. Is observed $\Delta O_3(\tau) > \text{instrumental error (comparison flights)}$
 $> \text{uncertainty in value at origins, } O_3(t_0 - \tau)$?
- 3. What is the sensitivity of ΔO_3 to initial conditions, reaction pathways, mixing history, ...?

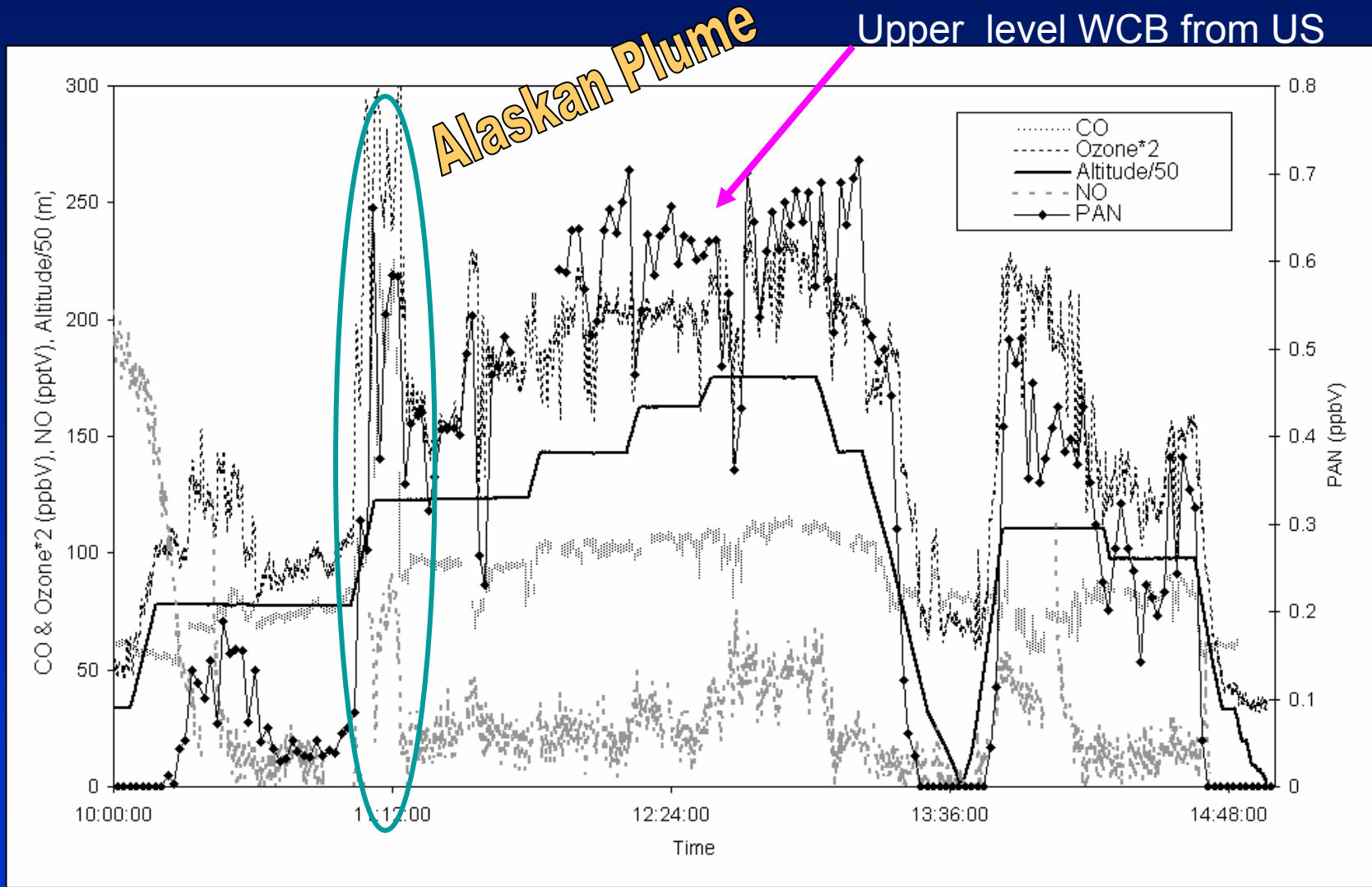
Alaskan Biomass Plumes: B032

PAN & CO were extremely elevated within the plume and displayed a strong, positive correlation
Ozone seemed somewhat suppressed within the plume



Alaskan biomass plume was extensively sampled at 5.6 km

In Contrast...B036



Plume Comparison

	B032 Plume	B036 Plume
PAN	1.72 ppbV	0.43 ppbV
O ₃	92 ppbV	133 ppbV
CO	261.2 ppbV	132.8 ppbV
(PAN/NO _y)*100	39 %	15 %
Temperature	263.4 K	268.0 K

Work in progress

Lagrangian matching using tracers + trajectories. (*Reading / Leeds / York*)

PAN / ozone model studies. (*Leeds / York*)

3D model diagnostics of ozone (*Cambridge*)

Box model / trajectory calculations of o-VOC production (*Leeds*)

HOx determinants (*Leeds*)

Aerosol composition analysis (*Manchester*)

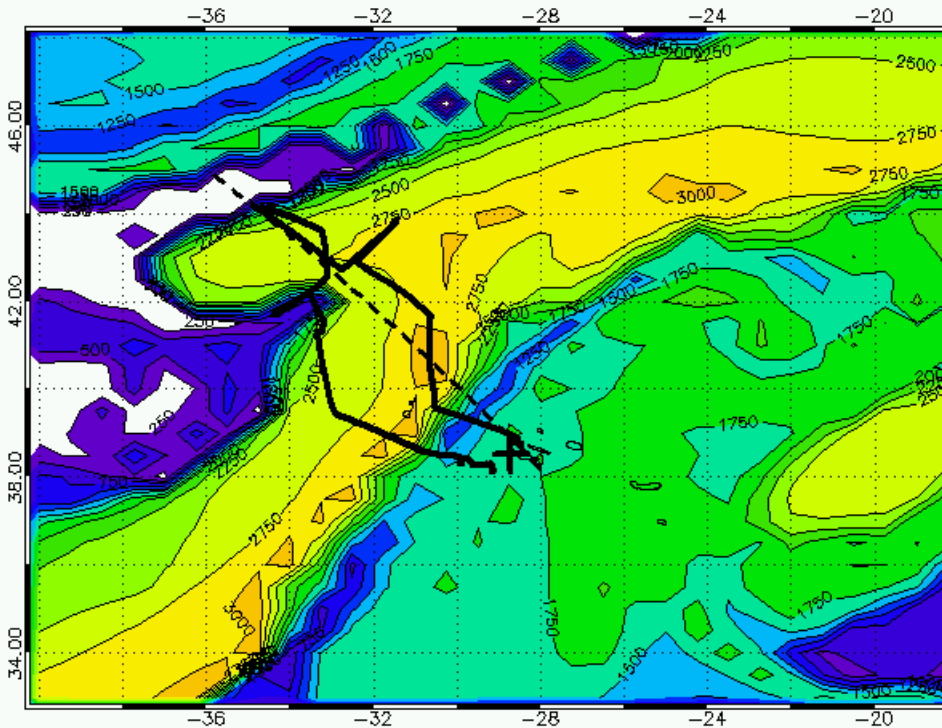
Lagrangian model to investigate sensitivity of trans-Atlantic ΔO_3 to:

- *Mixing rate*
- *Cloud cover*
- *ECMWF water vapour*
- *NMHC complexity*
- *Initial conditions*
- Constraint of mixing term through hydrocarbons.
- Full Monte-Carlo analysis using Lagrangian model.
- Can reduced chemical mechanisms describe the observed change in composition or is complexity unavoidable?

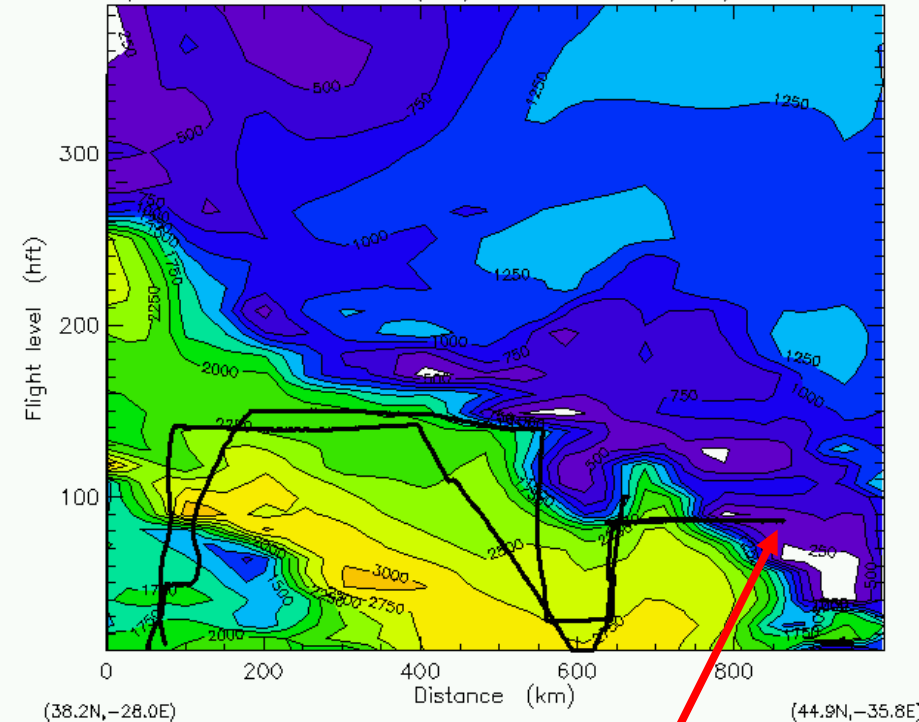
UK ITOP flight by flight summary

B029: 15/07/04 Low level pollution and fires

Intercept dist from base 1 (km) 850 hPa
Release on 12UT 15/07/2004 from 000hr met forecast



Intercept dist from base 1 (km) 12UT 15/07/2004 F+000

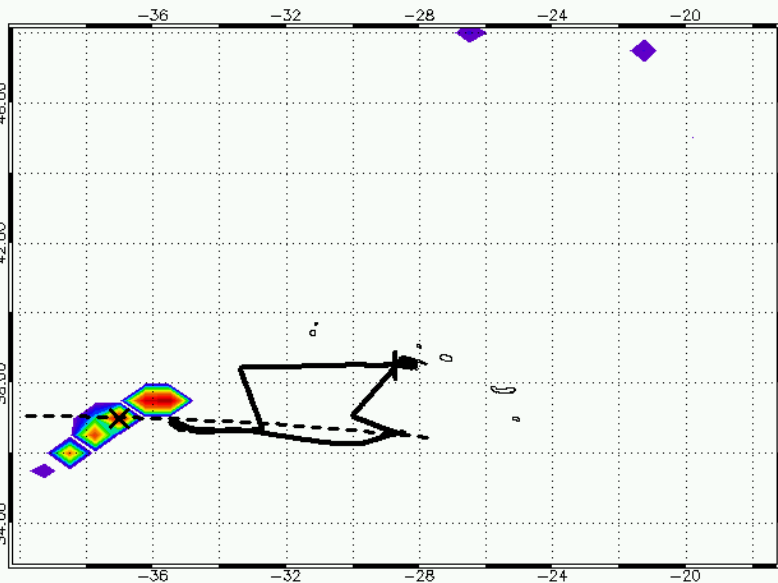


Horta

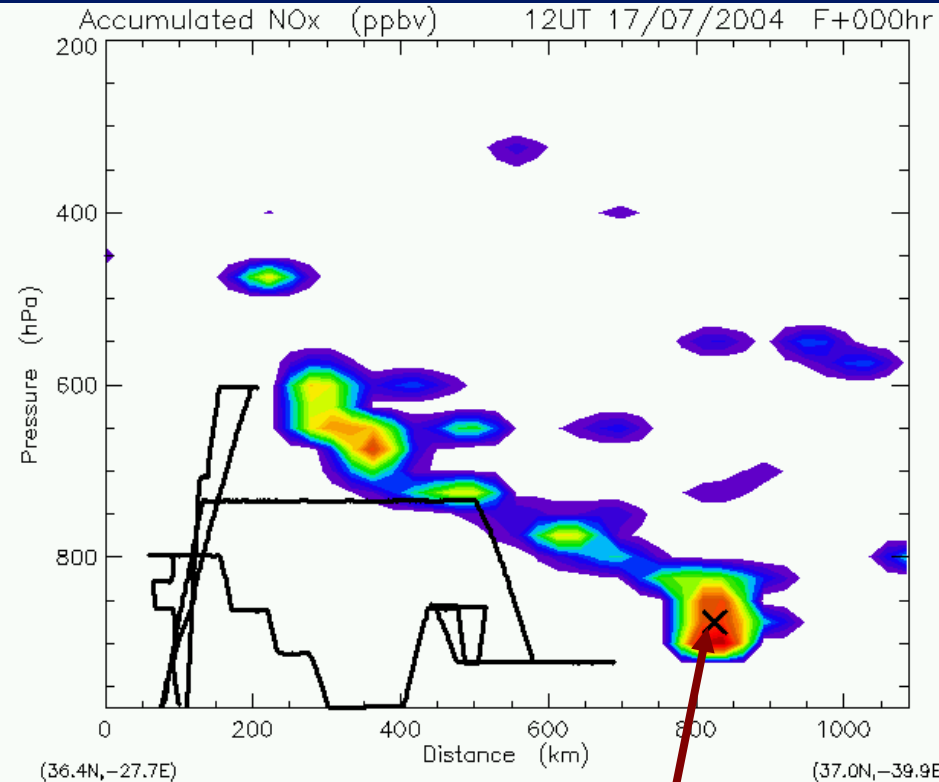
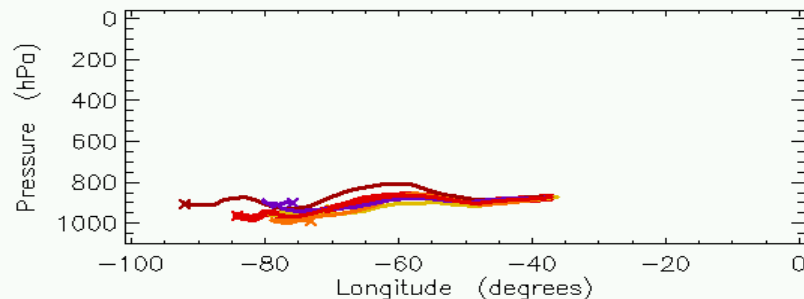
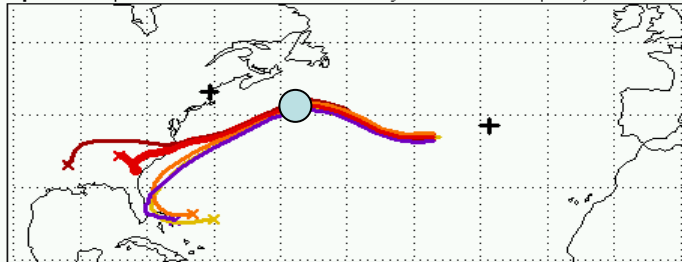
Air passing
close to Pease

B030: 17/07/04 First Lagrangian opportunity

Accumulated NO_x (ppbv) 875 hPa
Release on 12UT 17/07/2004 from 000hr met forecast



$\lambda = -37.0$, $\phi = 37.0$, $p = 875$ hPa, $T = -7.00$ days, 12UT 17/07/2004 F+000hr



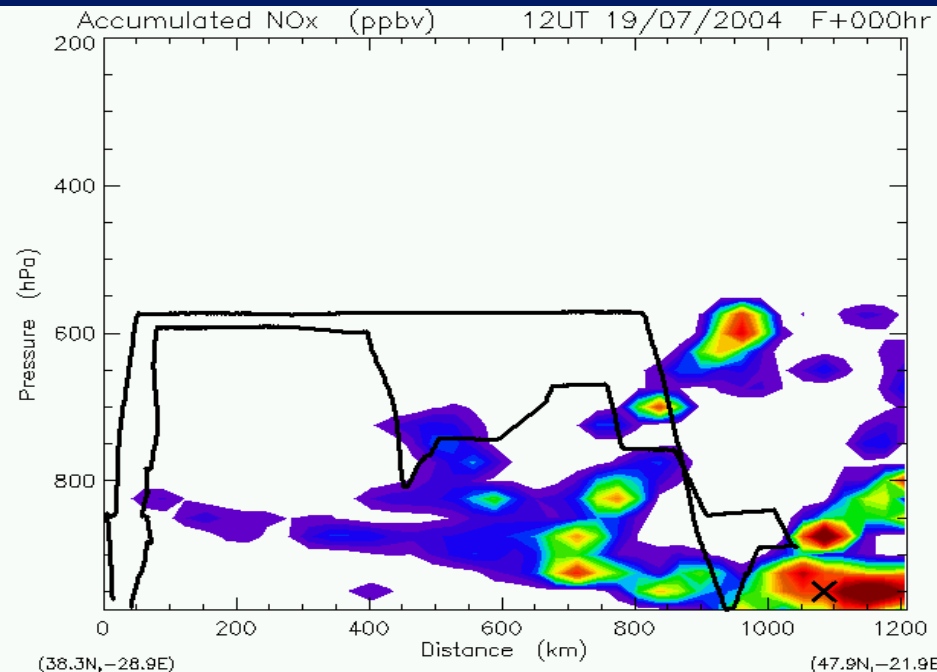
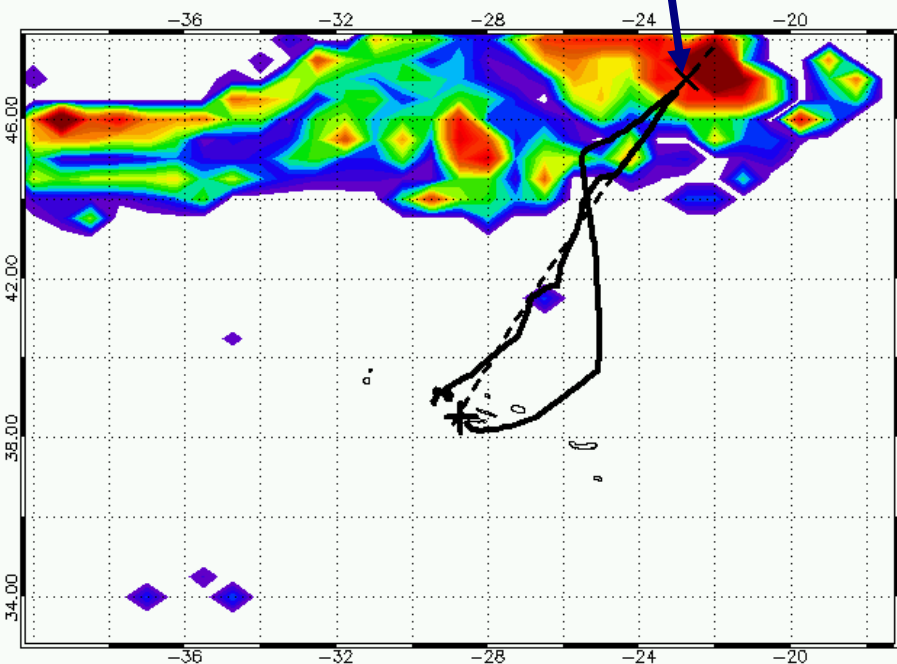
Target sampled by P3 on 15 July

Unfortunately only skimmed edge because out of time.

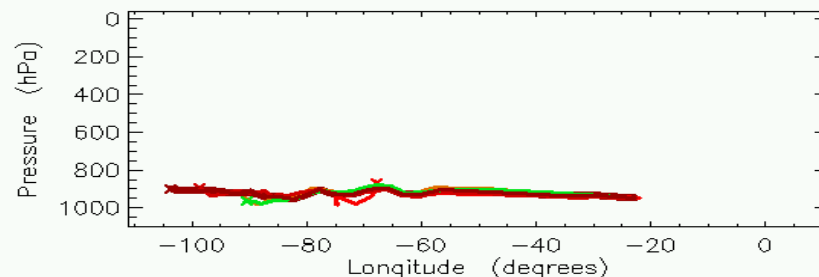
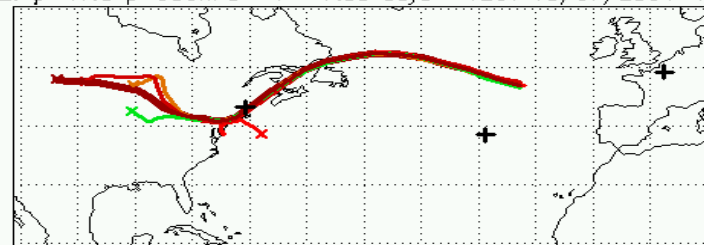
B031: 19/07/04 “New York plume”

Target sampled by P3 on 15 July (on limit of range)

Accumulated NO_x (ppbv) 950 hPa
Release on 12UT 19/07/2004 from 000hr met forecast

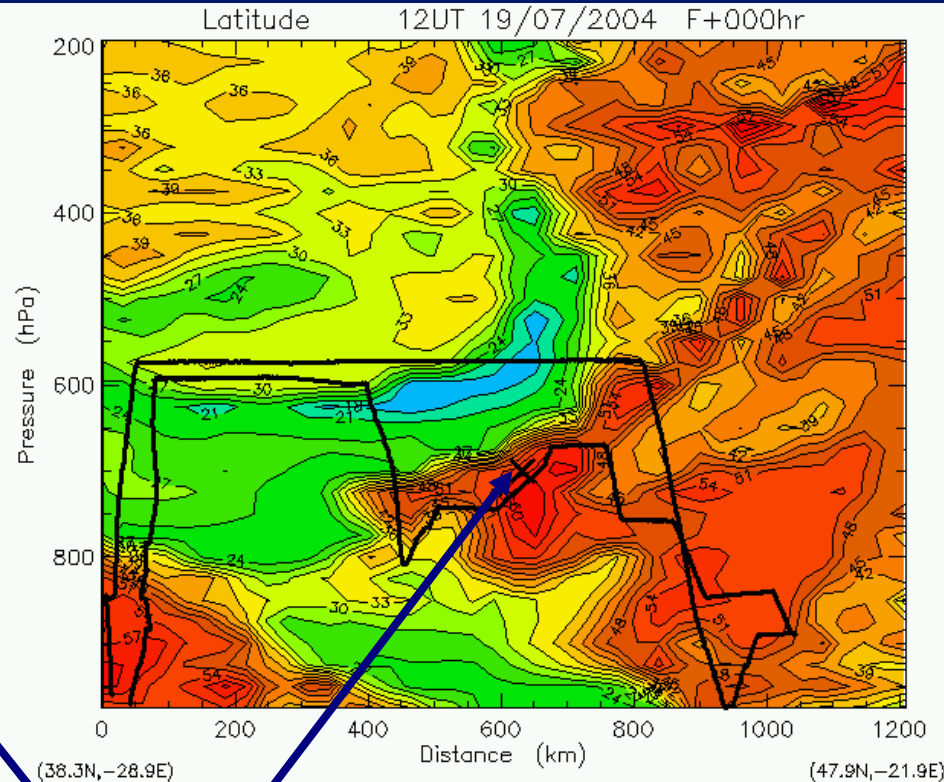
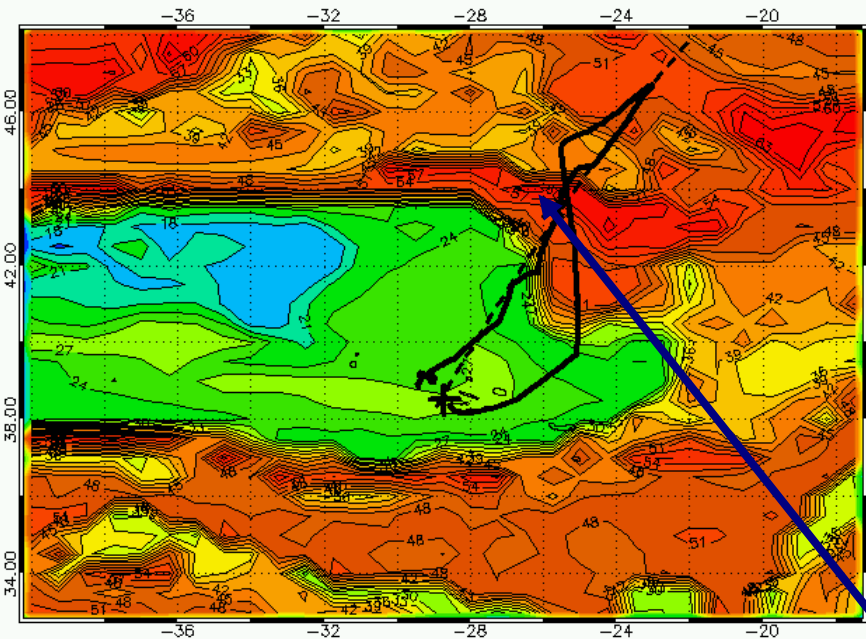


$\lambda = -22$, $\phi = 47.0$, $p = 950$ hPa, $T = -7.00$ days, 12UT 19/07/2004 F+000hr



B031 continued: thin brown fire layer

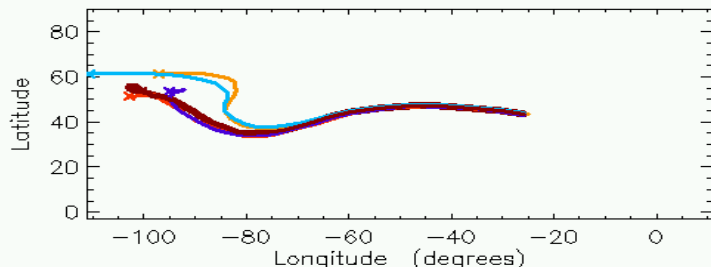
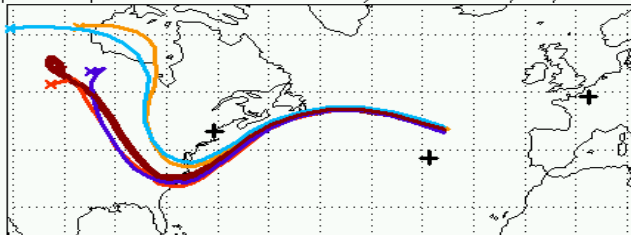
Latitude 700 hPa
Release on 12UT 19/07/2004 from 000hr met forecast



**Narrow filament and thin layer - visibly brown.
Adjusted altitude to stay within layer.**

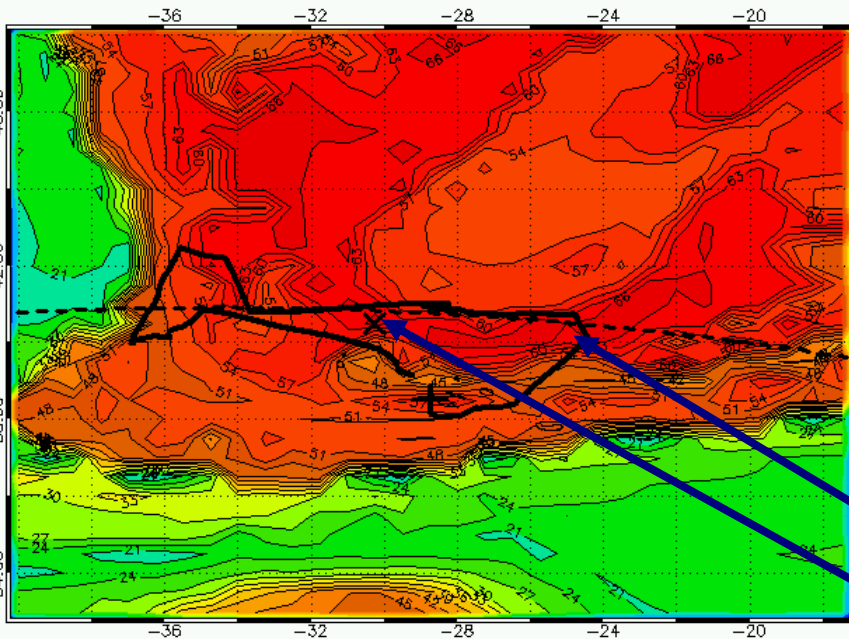
**Originated from UT above Alaskan forest fires
(but diluted by mixing while thinning).**

$\lambda = -25$, $\phi = 43.5$, $p = 700$ hPa, $T = -7.00$ days, 12UT 19/07/2004 F+000hr

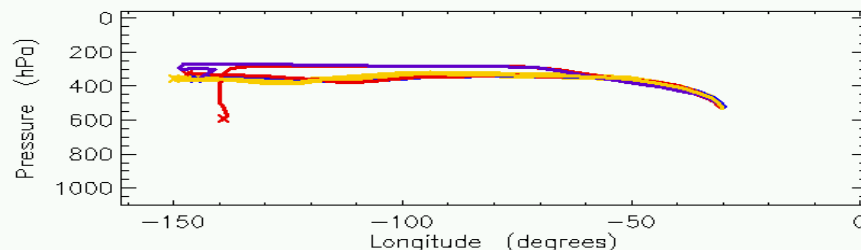
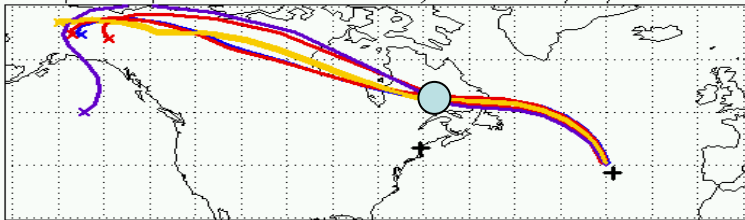


B032: 20/07/04 Alaskan forest fires

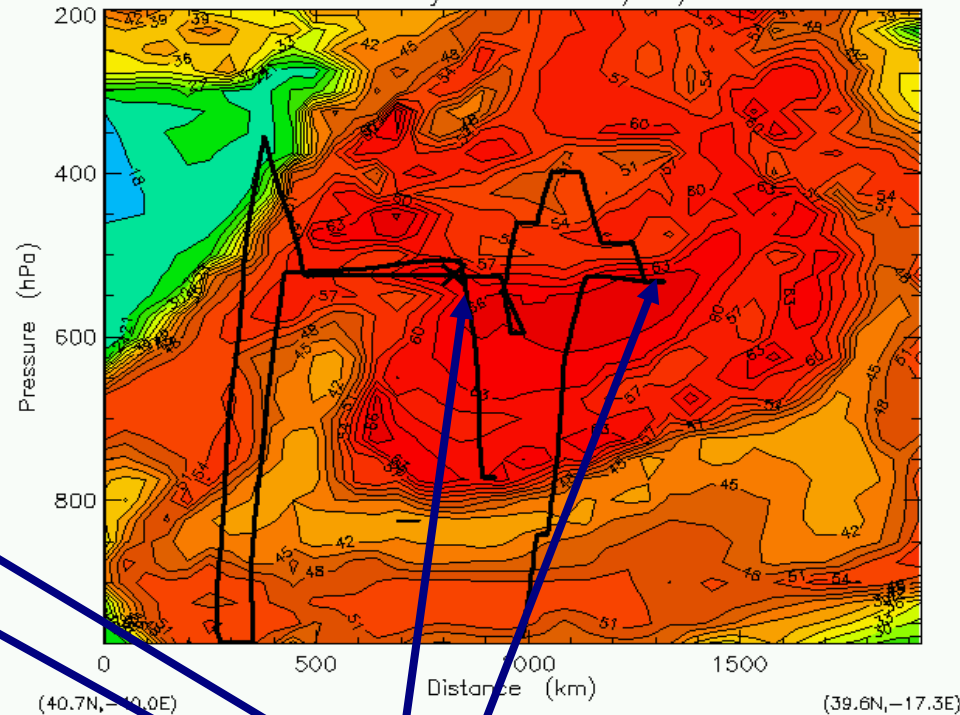
Latitude T=-6.00 days 525 hPa
Release on 12UT 20/07/2004 from 000hr met forecast



$\lambda = -30$, $\phi = 40.5$, $p = 525$ hPa, T=-6.00 days 12UT 20/07/2004 F+000hr



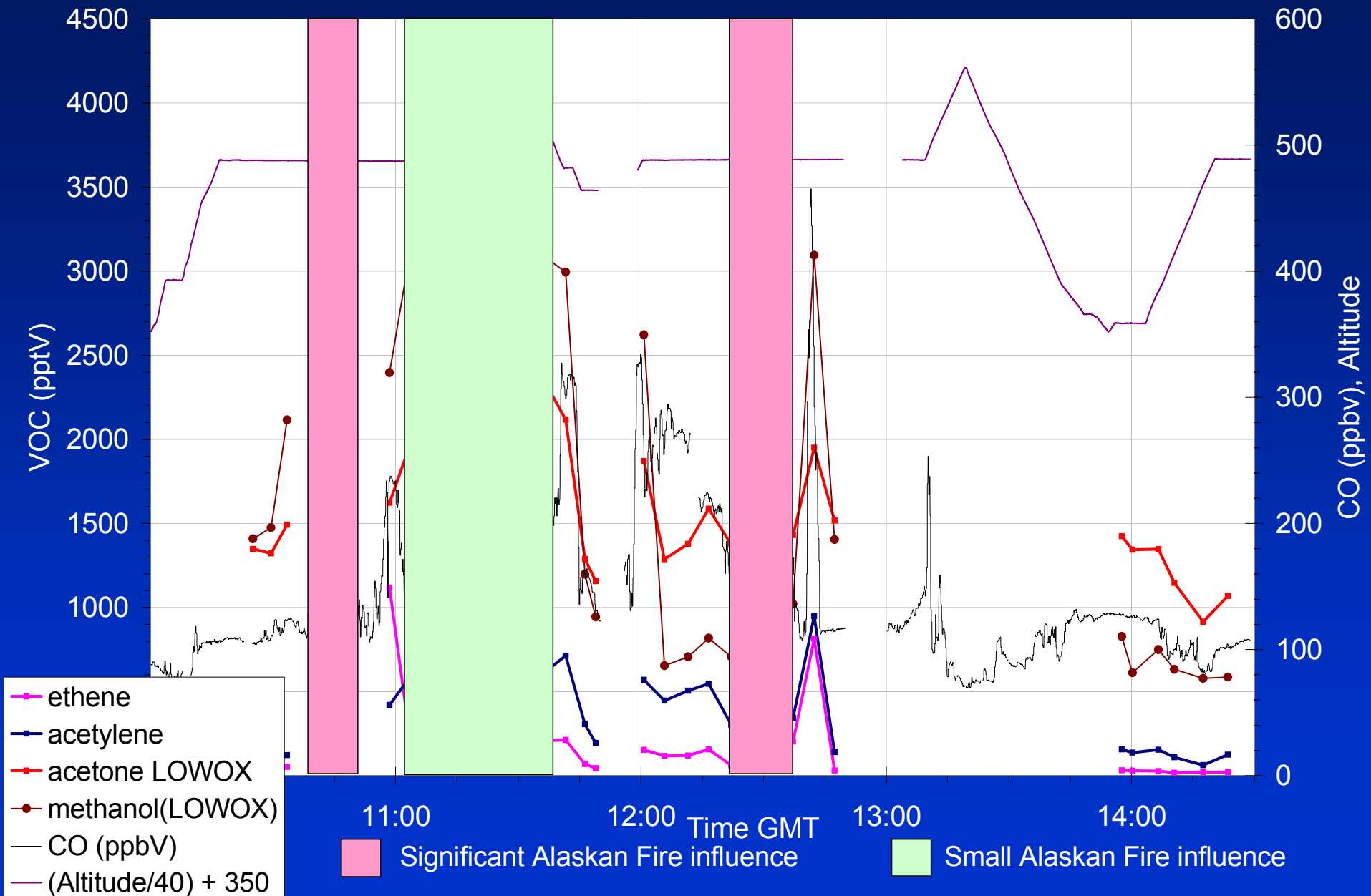
Latitude T=-6.00 days 12UT 20/07/2004 F+000hr



Alaskan fire plume with CO up to 600ppb.
Similar plume was intercepted by P3 on 15
July (but not same air mass).

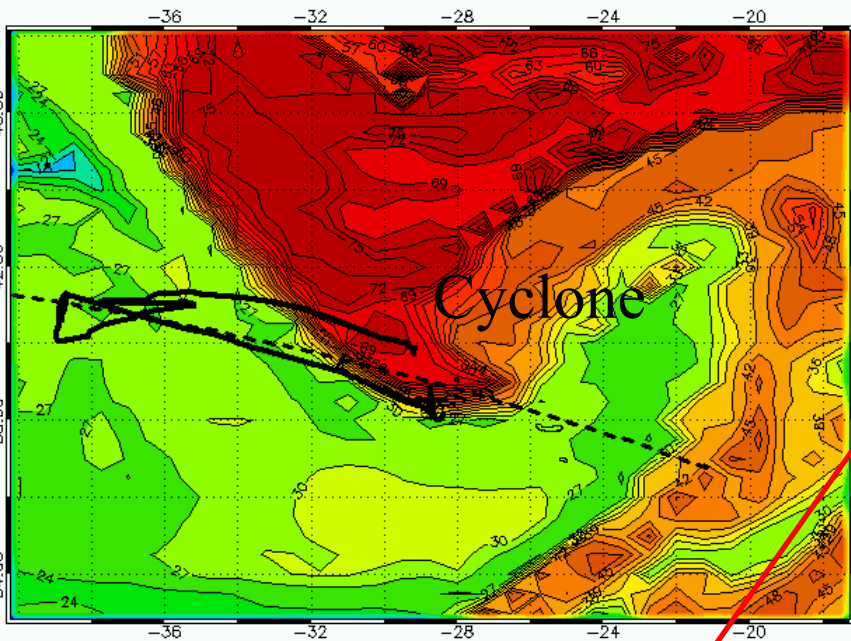
Was targeted on 23/7 by Falcon.

B032 NHMCs

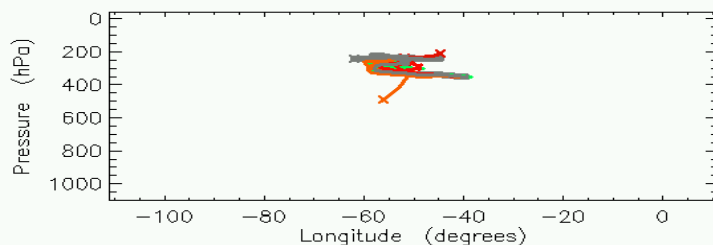
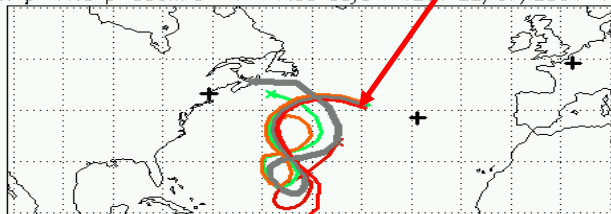


B033: 22/07/04 ENVISAT underpass

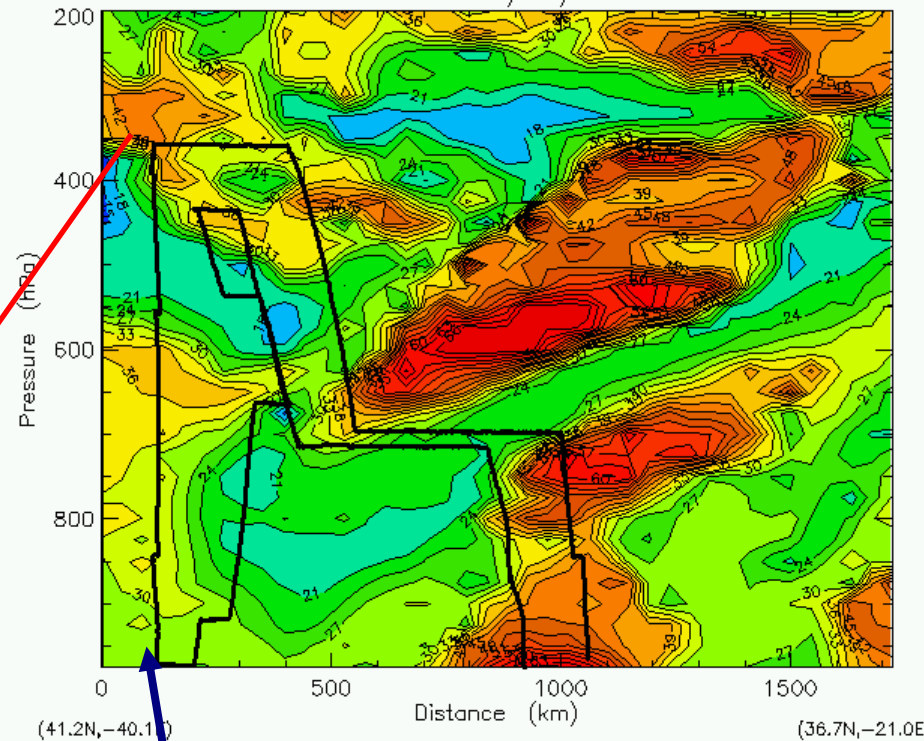
Latitude 975 hPa
Release on 12UT 22/07/2004 from 000hr met forecast



$\lambda = -39$, $\phi = 41.0$, $p = 350$ hPa, $T = -7.00$ days, 12UT 22/07/2004 F+000hr



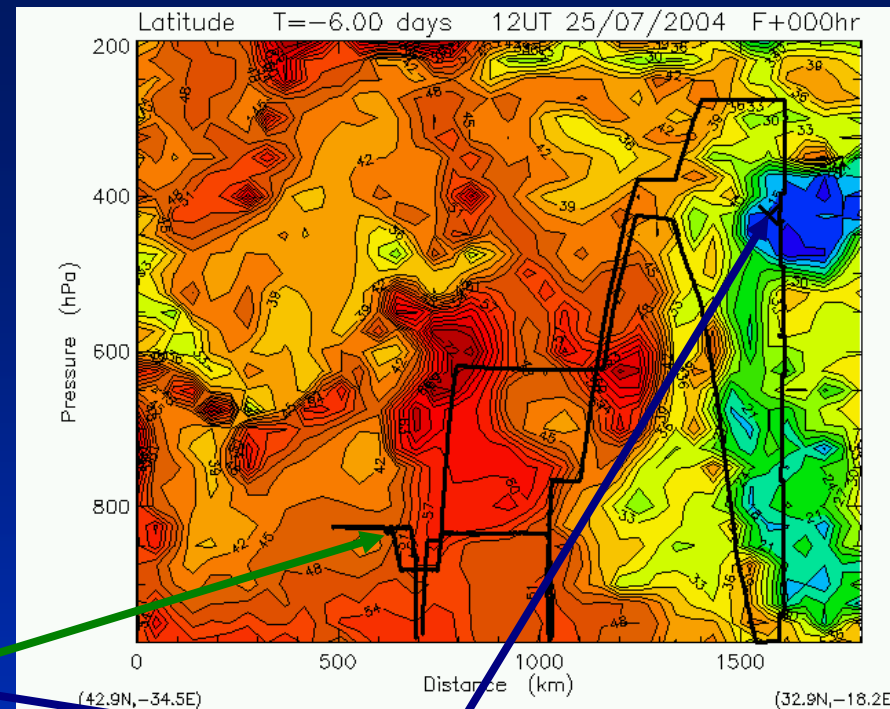
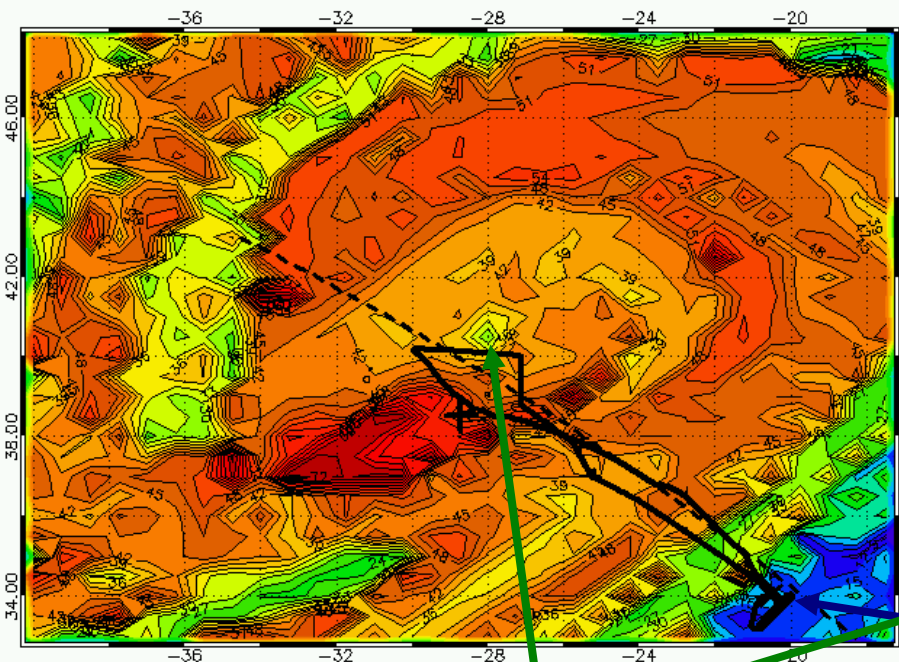
Latitude 12UT 22/07/2004 F+000hr



Deep profile under satellite
through air re-circulating over
mid-Atlantic for more than a
week.

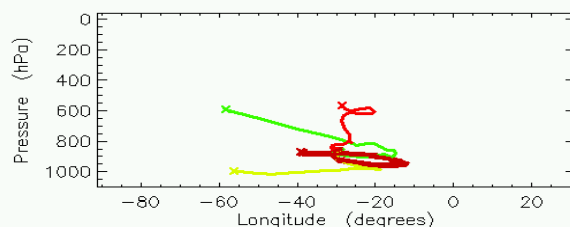
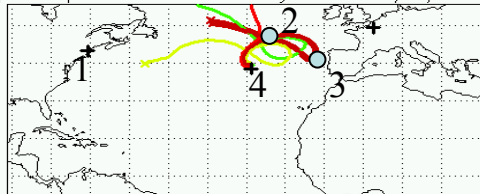
B034: 25/07/04 African air

Latitude T=-6.00 days 425 hPa
Release on 12UT 25/07/2004 from 000hr met forecast



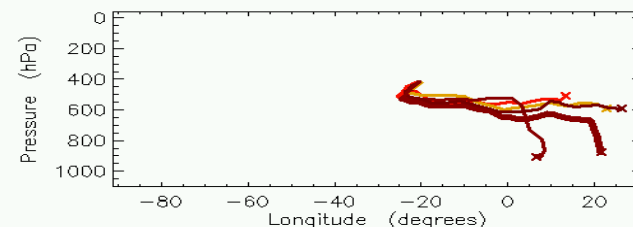
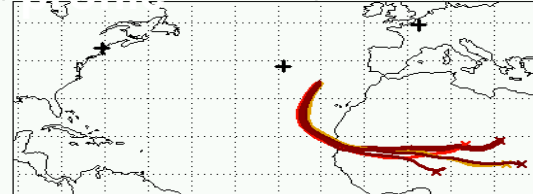
New York plume 4th interception

$\lambda = -28$, $\phi = 39.5$, $p = 850$ hPa T=-7.00 days 12UT 25/07/2004 F+000hr



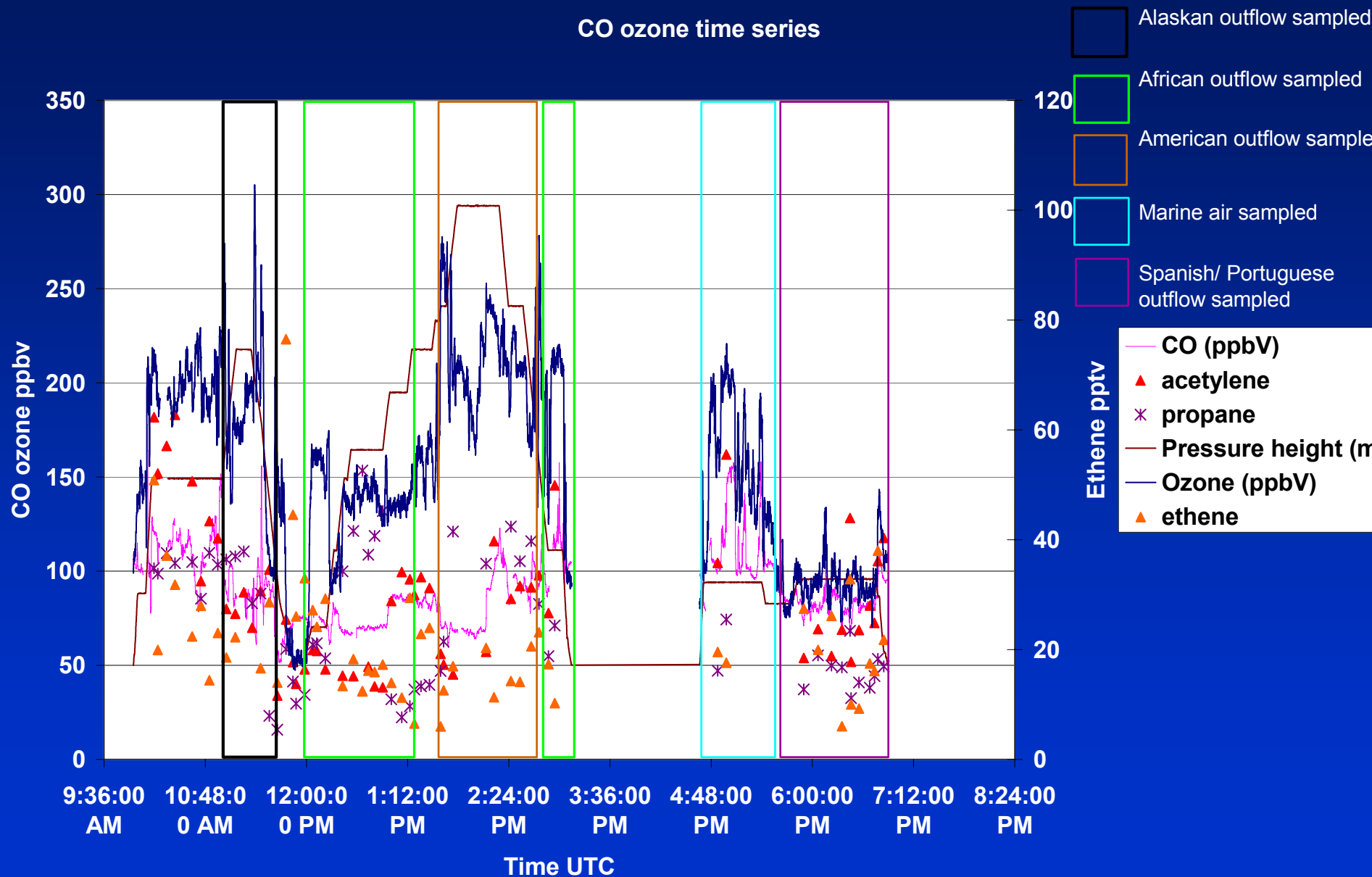
African air throughout

$\lambda = -20$, $\phi = 39.5$, $p = 850$ hPa T=-6.00 days 12UT 25/07/2004 F+000hr



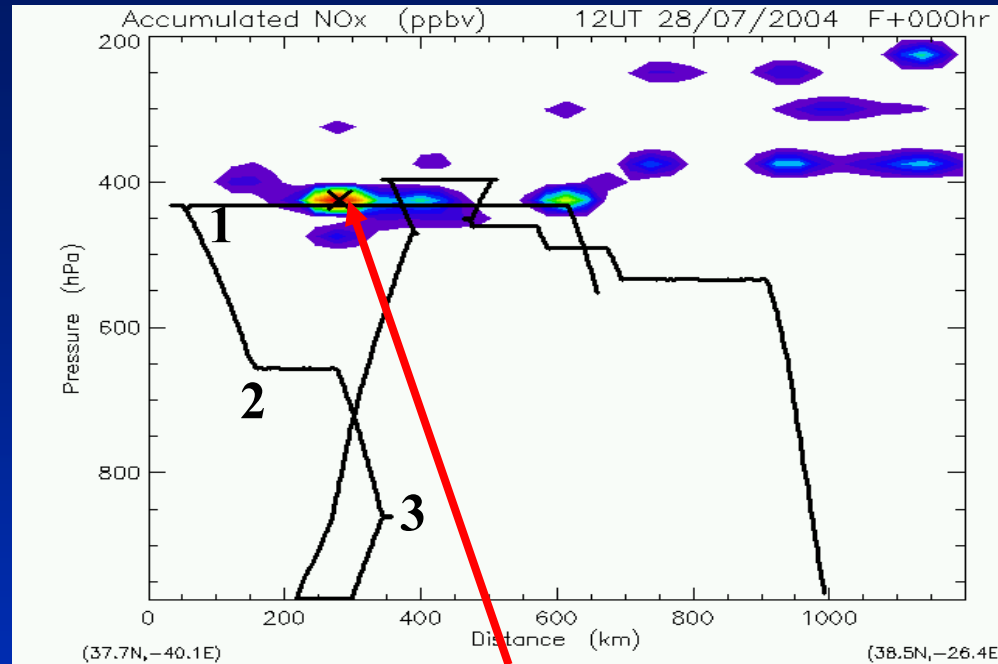
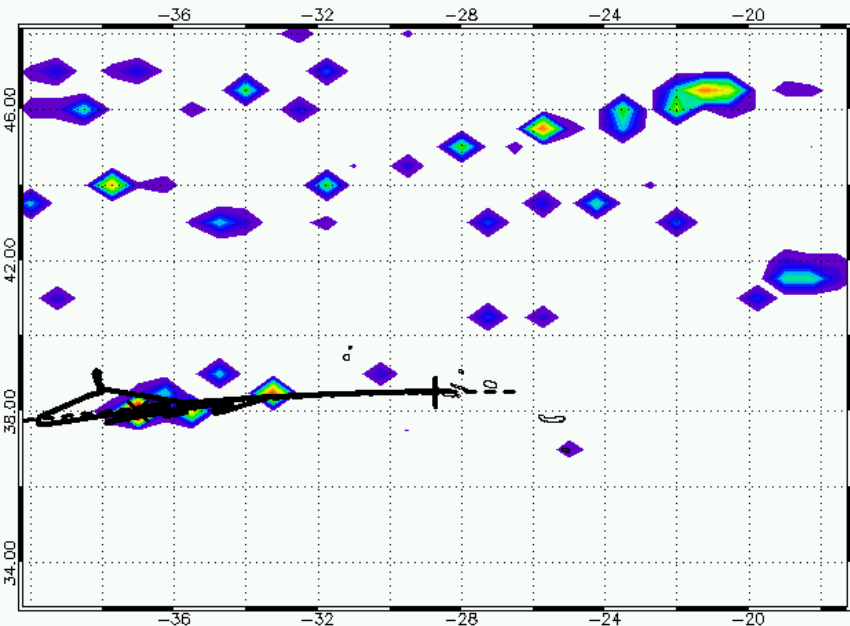
B034: 25/07/04 African air

CO ozone time series



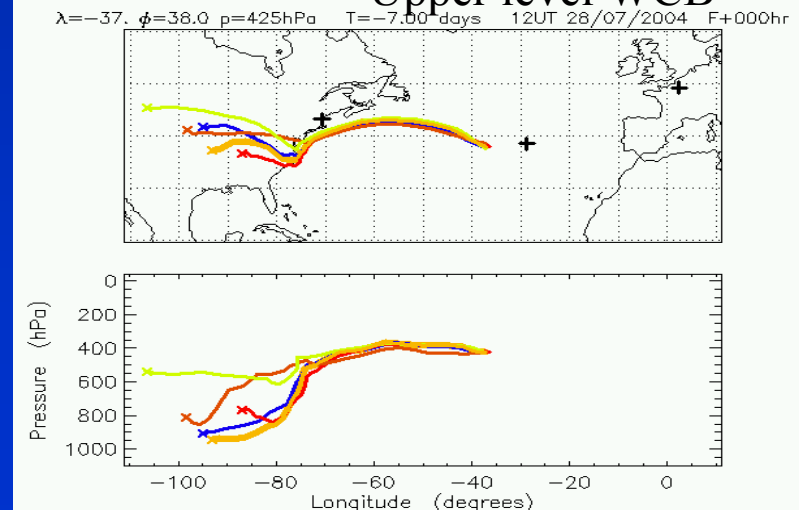
B035: 28/07/04 Comparison with DC8

Accumulated NO_x (ppbv) 425 hPa
Release on 12UT 28/07/2004 from 000hr met forecast



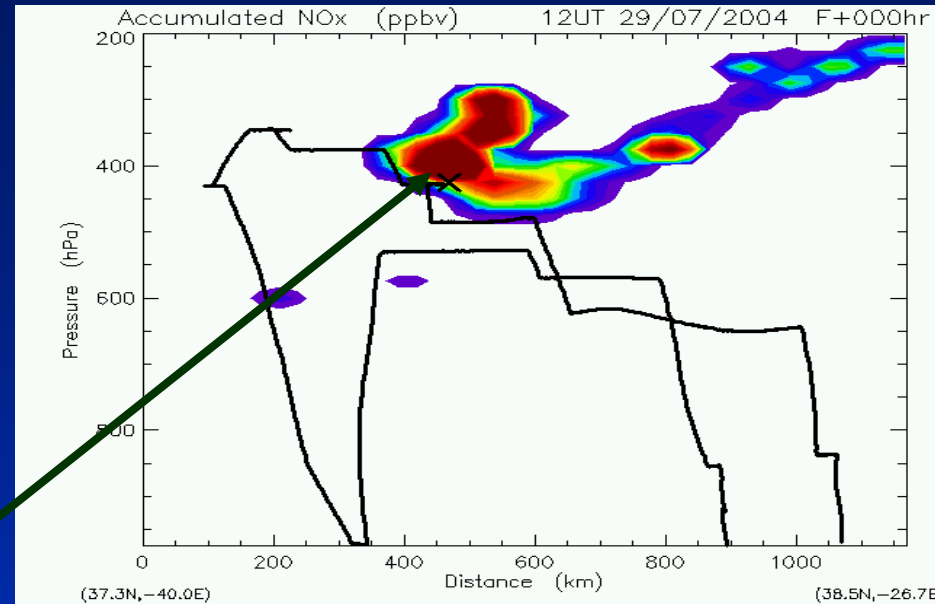
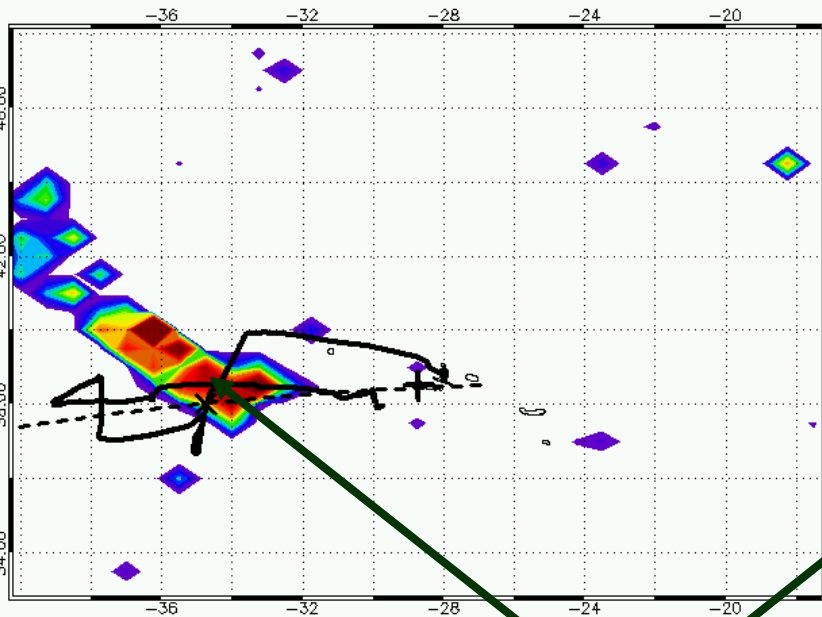
Comparison below air exported
by WCB. Relatively clean but
sampled again by Falcon on 30/7
and 31/7.

Upper level WCB



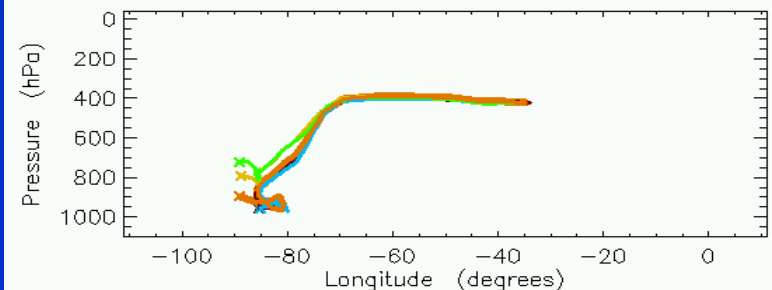
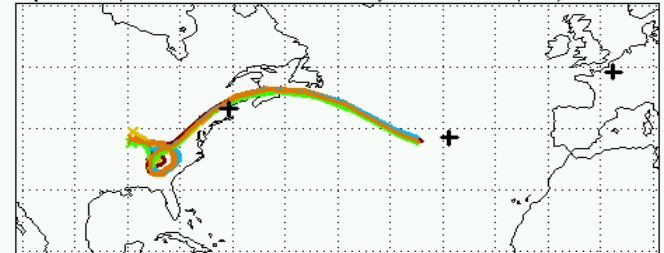
B036: 29/07/04 Upper level export in WCB

Accumulated NO_x (ppbv) 425 hPa
Release on 12UT 29/07/2004 from 000hr met forecast



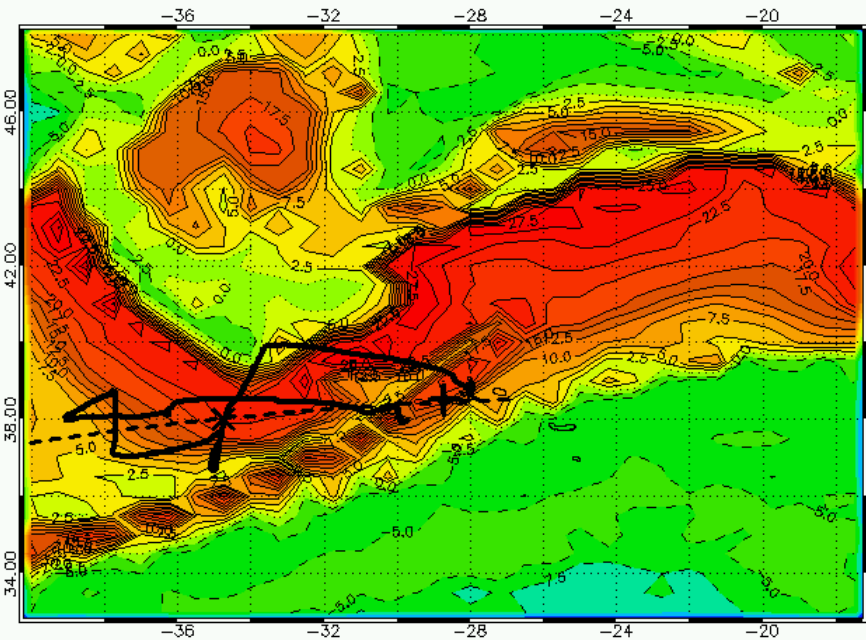
Polluted air on North side of WCB but crossed under maximum because could not achieve altitude early enough.

$\lambda = -34$, $\phi = 38.0$, $p = 425$ hPa, $T = -7.00$ days 12UT 29/07/2004 F+000hr



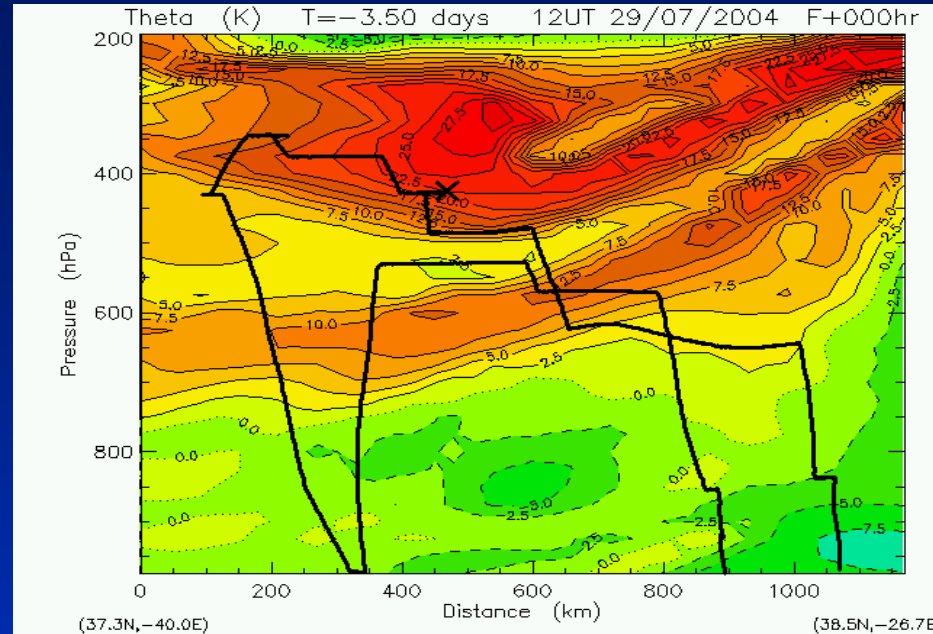
B036 continued: WCB features

Theta (K) T=-3.50 days 425 hPa
Release on 12UT 29/07/2004 from 000hr met forecast

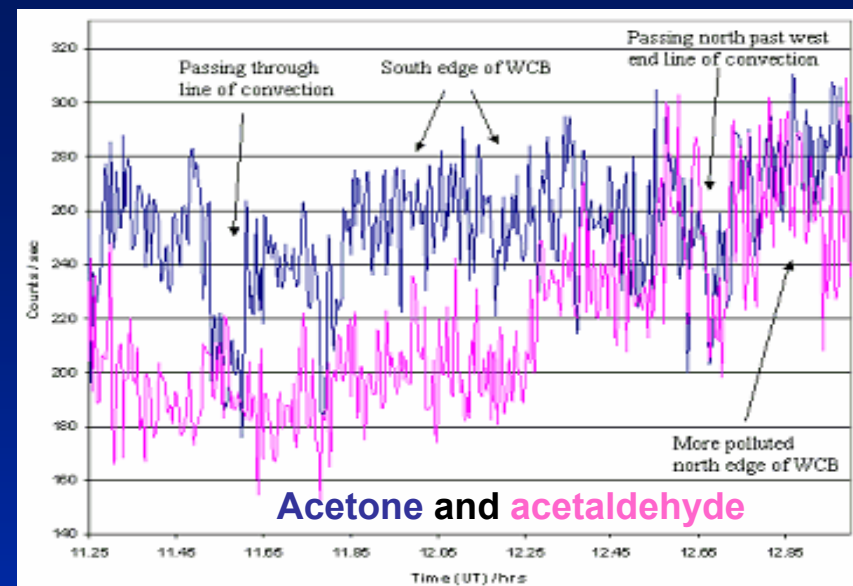
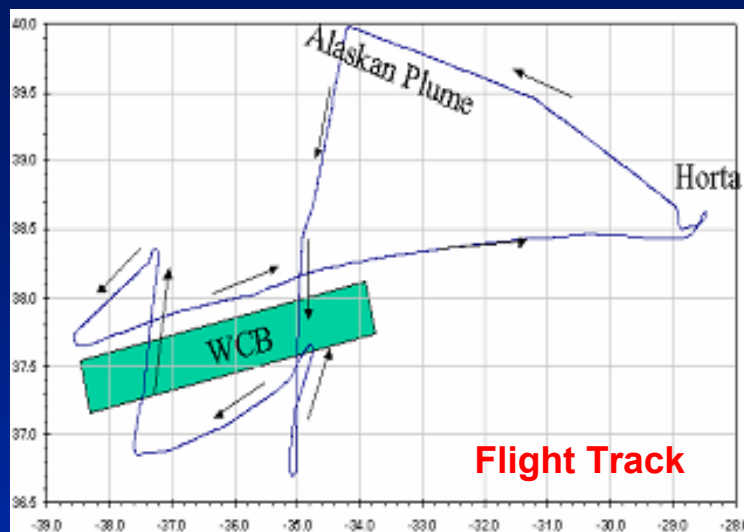


Observed line of convection
(dashed line) was parallel to
feature lying to south at upper
levels (figure above)

Narrow line of deep convection
– BA146 flew along south side
and then a deep profile on north
side



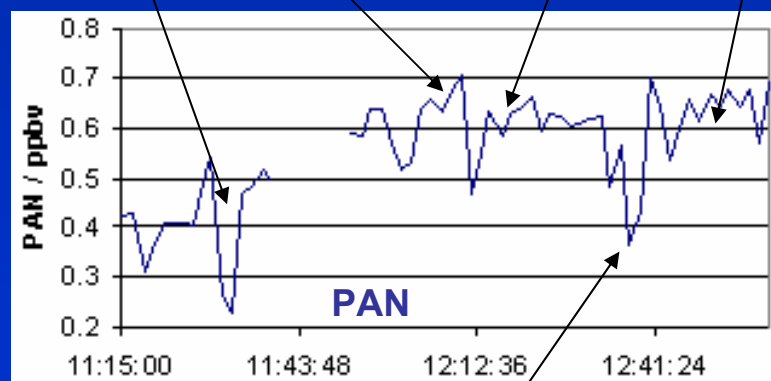
Flight B036



Through Cu-Nim gap

Along northern edge of WCB

Along southern edge of WCB

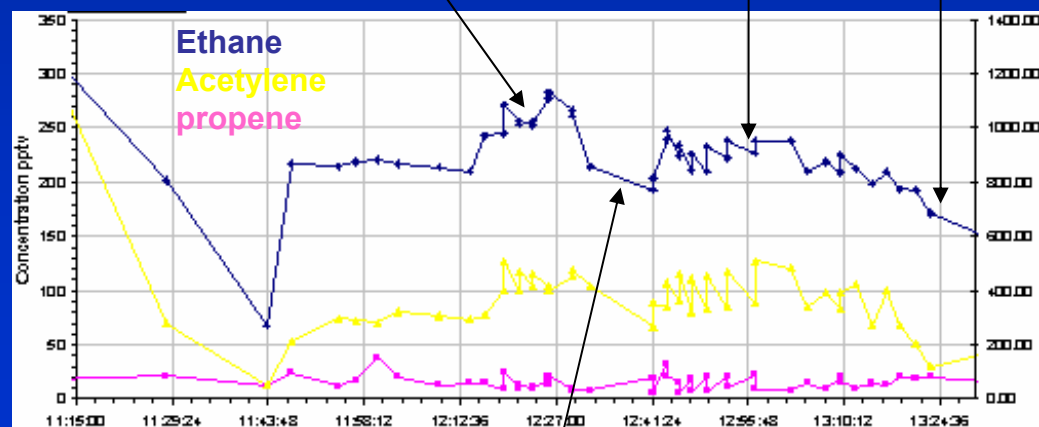


Passing north through western end of line of convection

Bottom of WCB outflow at FL180-160.

Southern edge of WCB

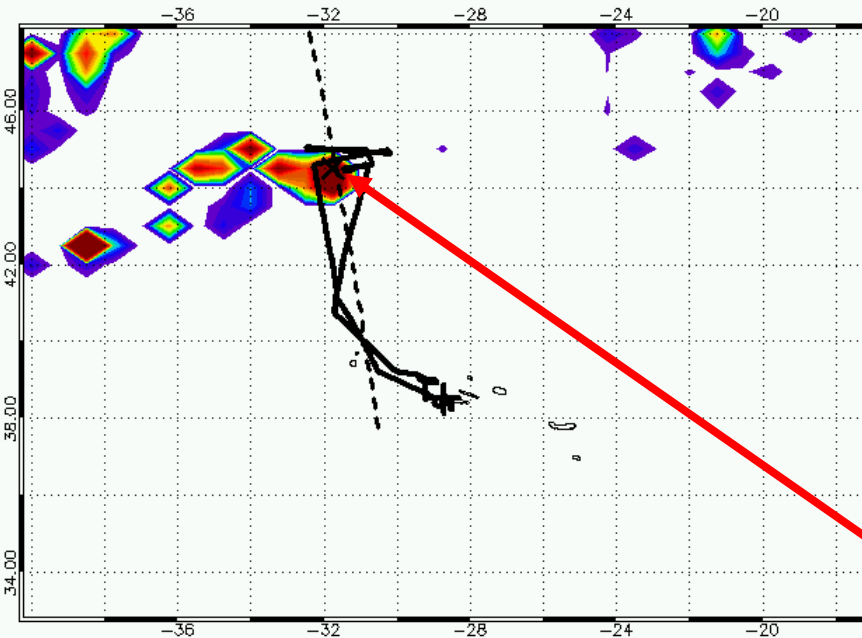
North of WCB



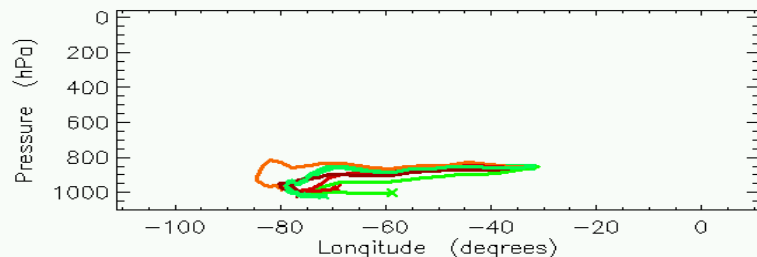
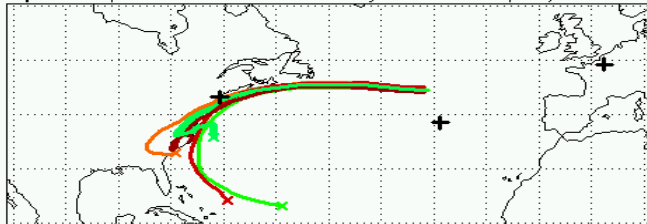
Crossing line of convection at western edge

B037: 31/07/04 Low level WCB (1)

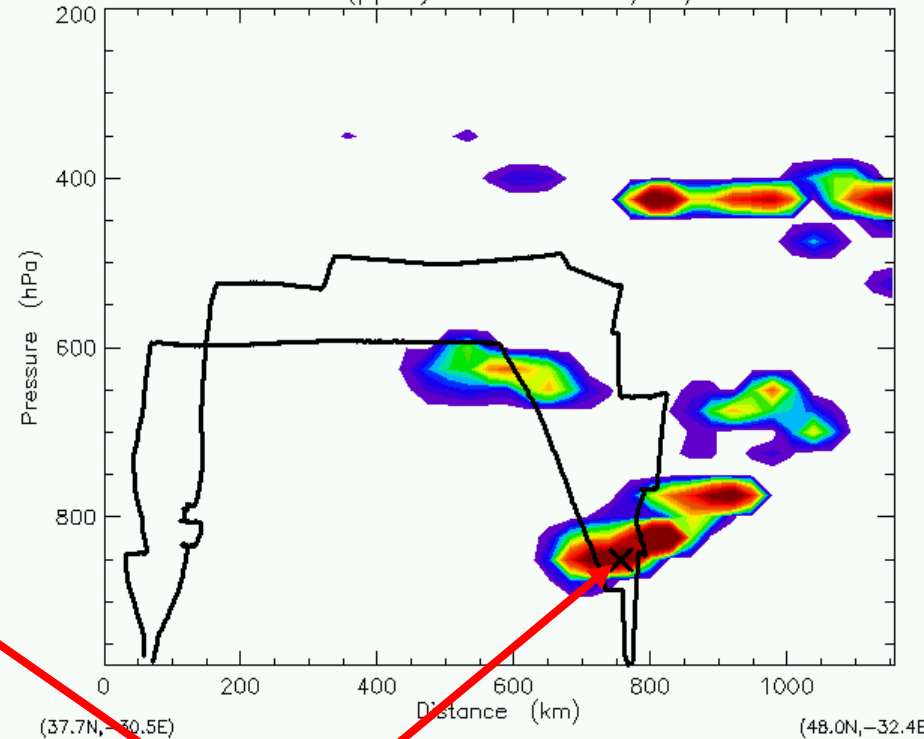
Accumulated NO_x (ppbv) 850 hPa
Release on 12UT 31/07/2004 from 000hr met forecast



$\lambda = -31$, $\phi = 44.5$, $p = 850$ hPa, $T = -7.00$ days, 12UT 31/07/2004 F+000hr



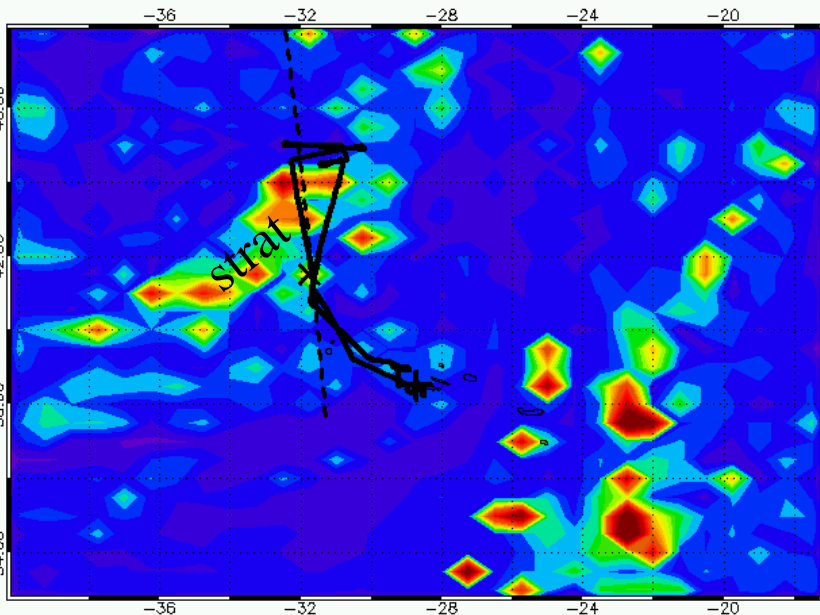
Accumulated NO_x (ppbv) 12UT 31/07/2004 F+000hr



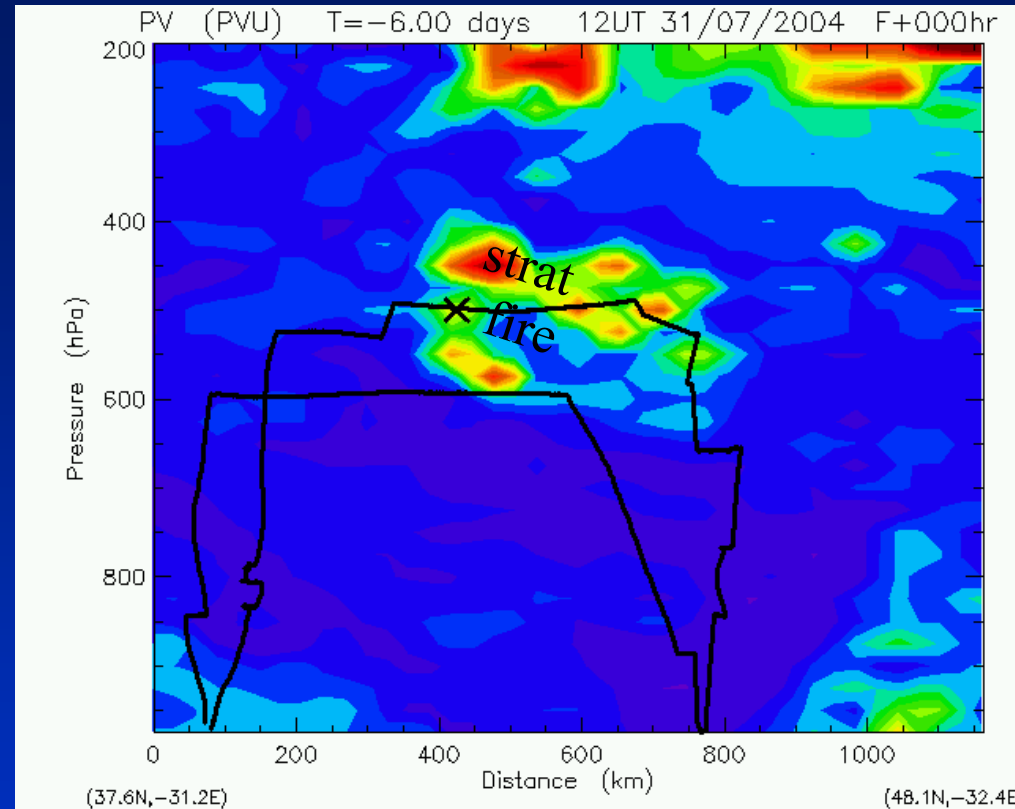
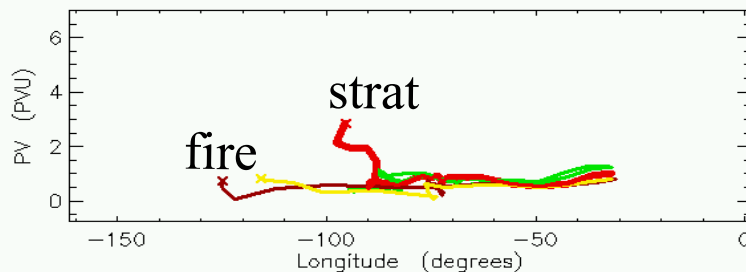
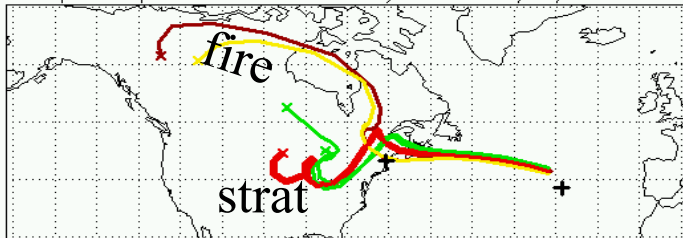
Target already sampled by
P3 on 27 and 28 July

B037 continued: forest fire mixed with stratospheric air

PV (PVU) T=-6.00 days 500 hPa
Release on 12UT 31/07/2004 from 000hr met forecast



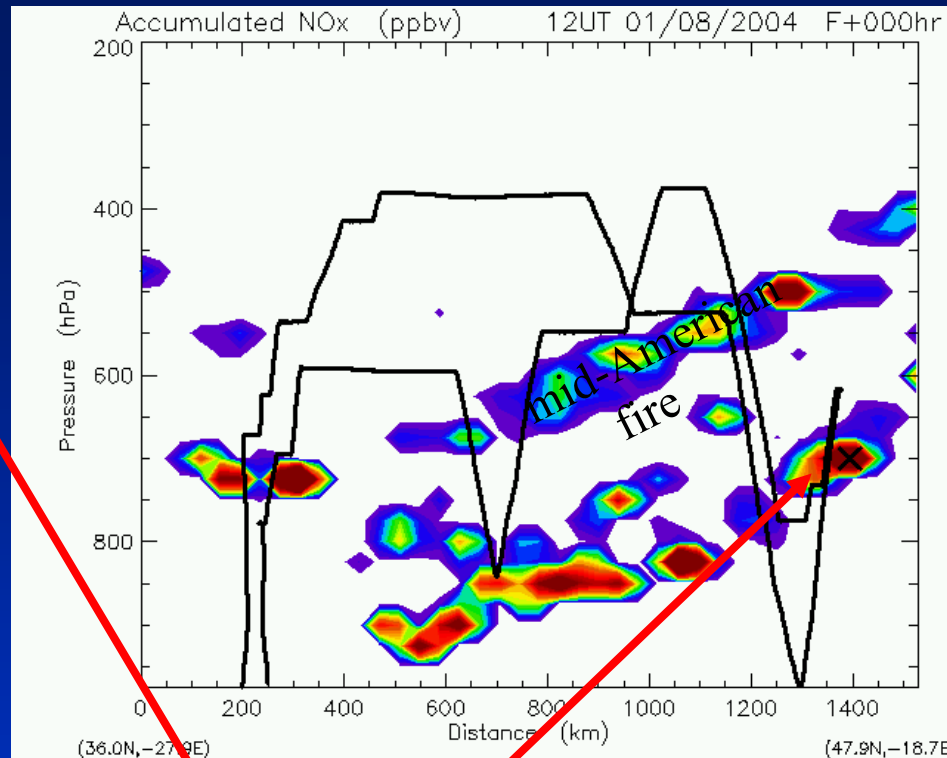
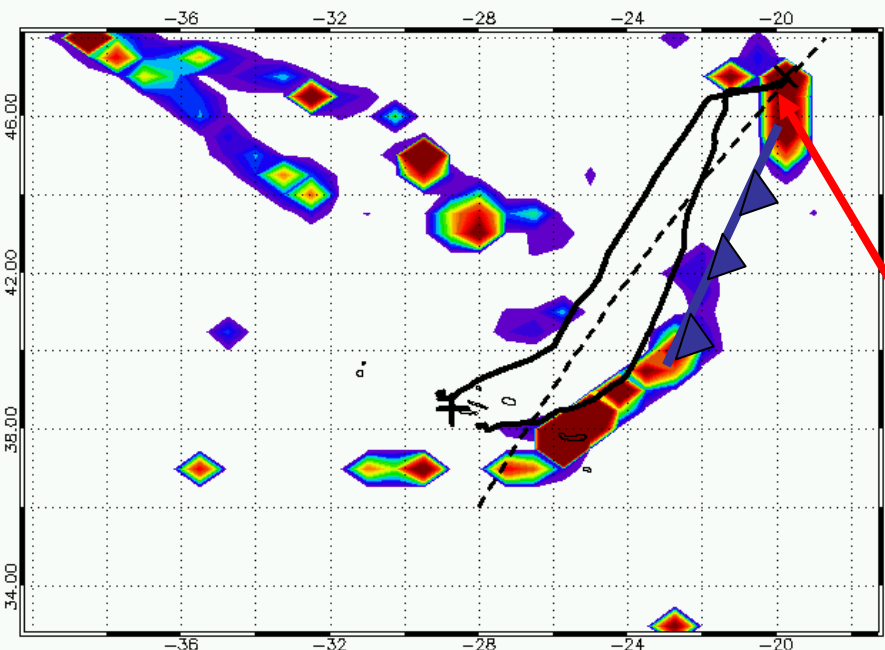
$\lambda = -31$, $\phi = 41.5$ p=500hPa T=-6.00 days 12UT 31/07/2004 F+000hr



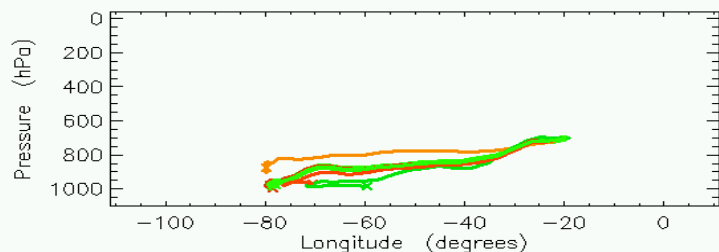
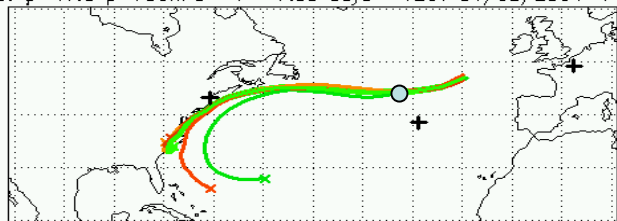
**Strong evidence for forest fire
CO mixed with stratospheric
ozone**

B038: 01/08/04 Low level WCB (2)

Accumulated NOx (ppbv) 700 hPa
Release on 12UT 01/08/2004 from 000hr met forecast

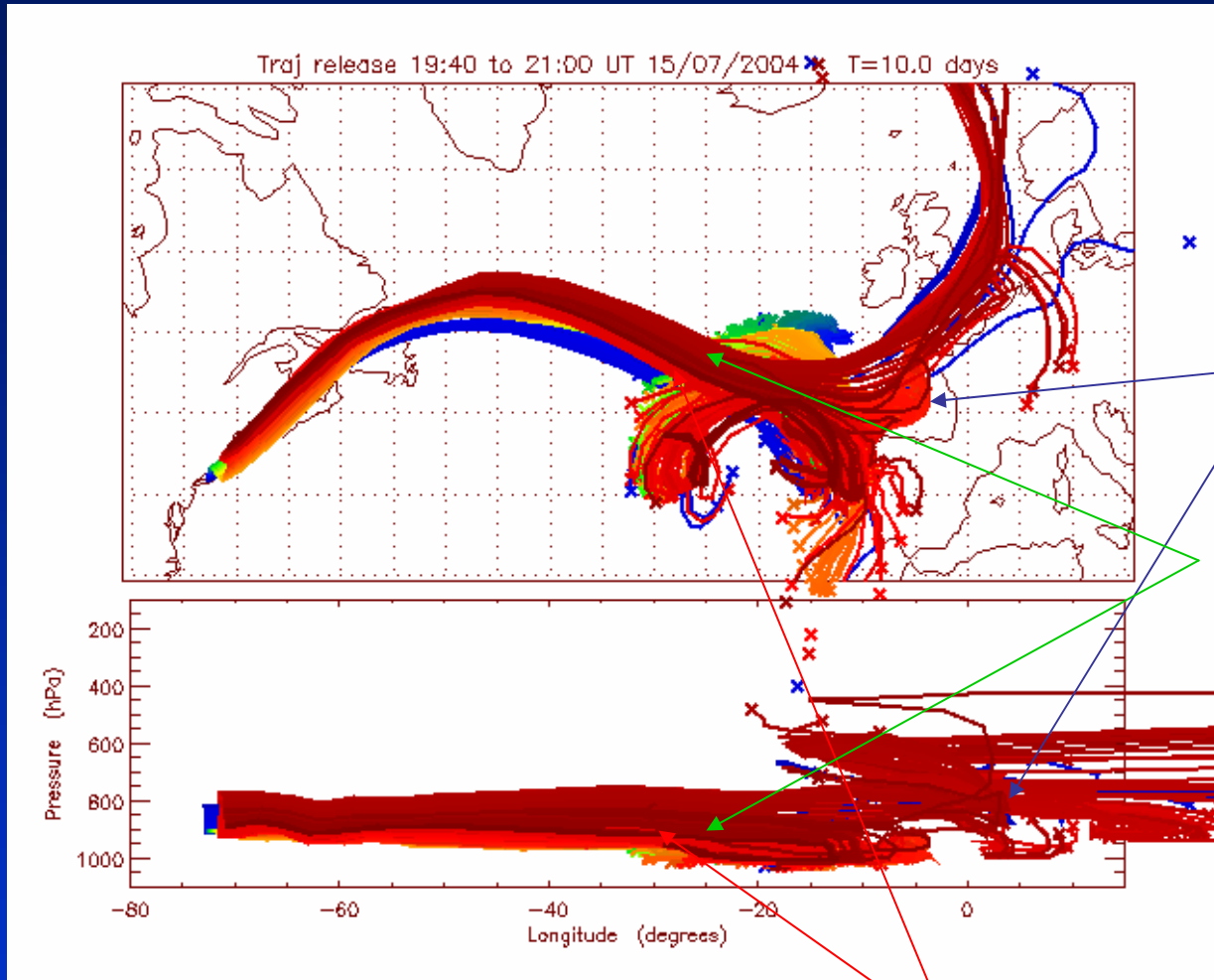


$\lambda = -19.0$, $\phi = 47.0$, $p = 700$ hPa, $T = -7.00$ days 12UT 01/08/2004 F+000hr



**Rapidly receding target
already sampled by P3 on 27
and 28 July and BAe146 on 31
July.**

Possible multiple interception of New York polluted airmass: 1



Intercepted by DLR Falcon
off NW corner of Spain on 22/7
(saw CO ~ 90-100ppb).

BAE146 flight intercepted this air
again on 25/7
(saw increased CO ~ 115-125ppb).

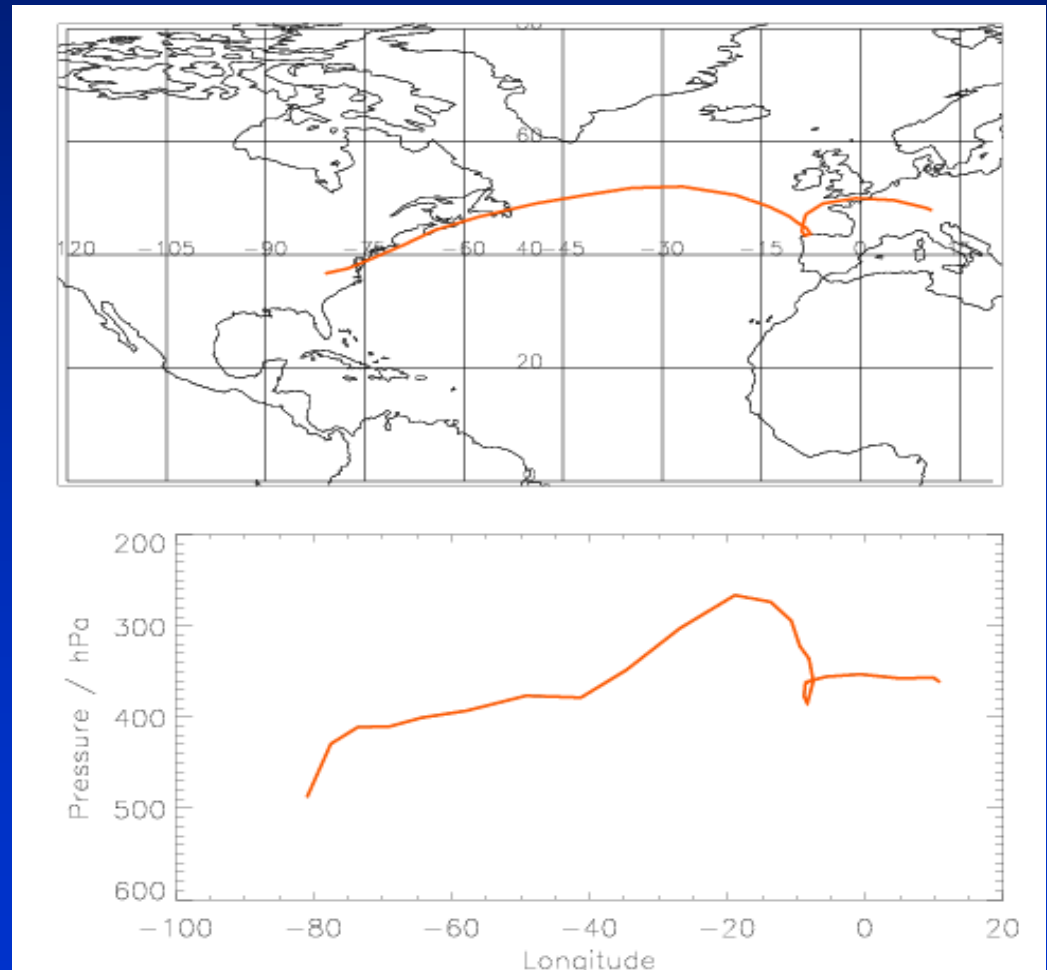
10 day forward trajectories from NOAA P3
flight off NYC on 15/7. Airmass doubles
back from Spain to the Azores.

BAE146 flight intercepted this air on 19/7
(saw CO ~ 115-125ppb).

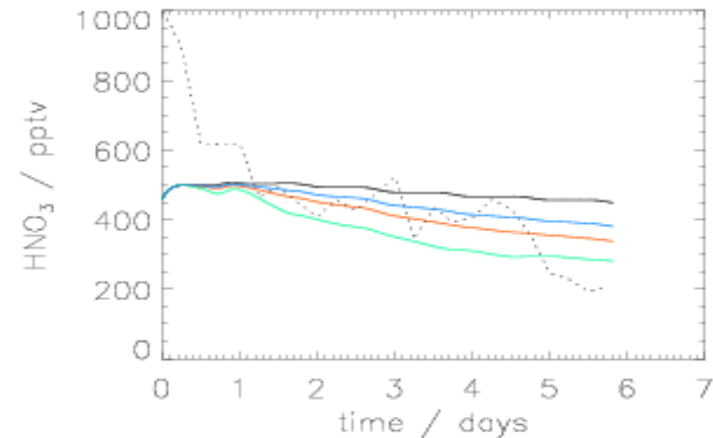
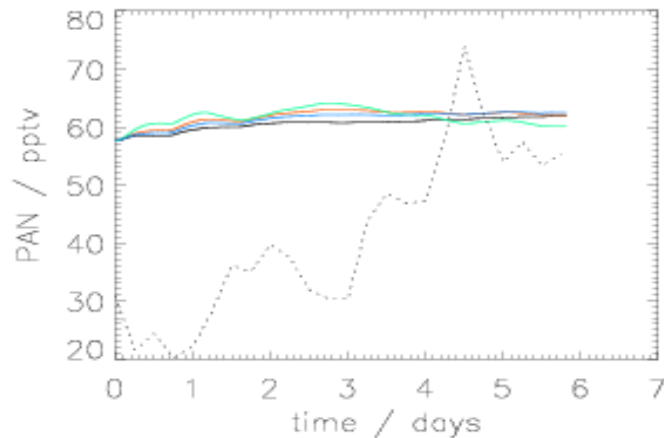
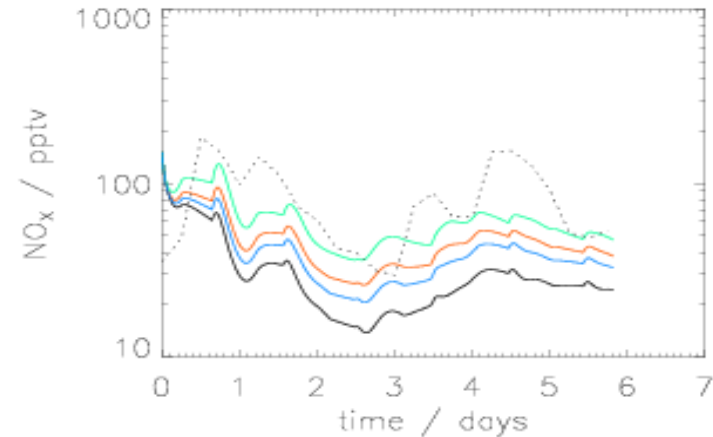
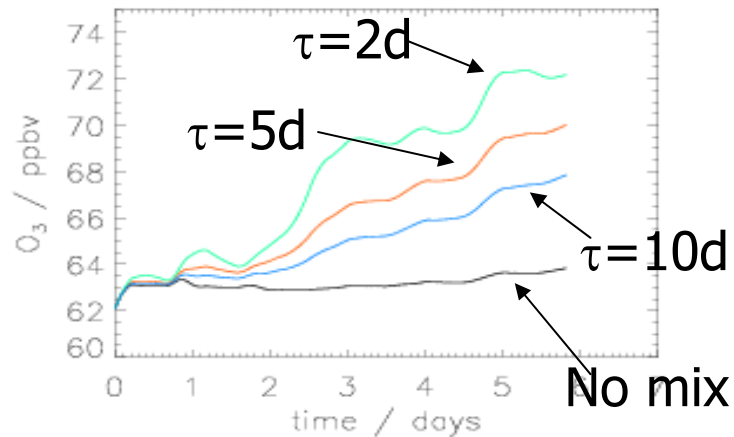
Sensitivity Analysis

Use Lagrangian model to investigate sensitivity of trans-Atlantic ΔO_3 to:

- Mixing rate
- Cloud cover
- ECMWF water vapour
- NMHC complexity
- Initial conditions

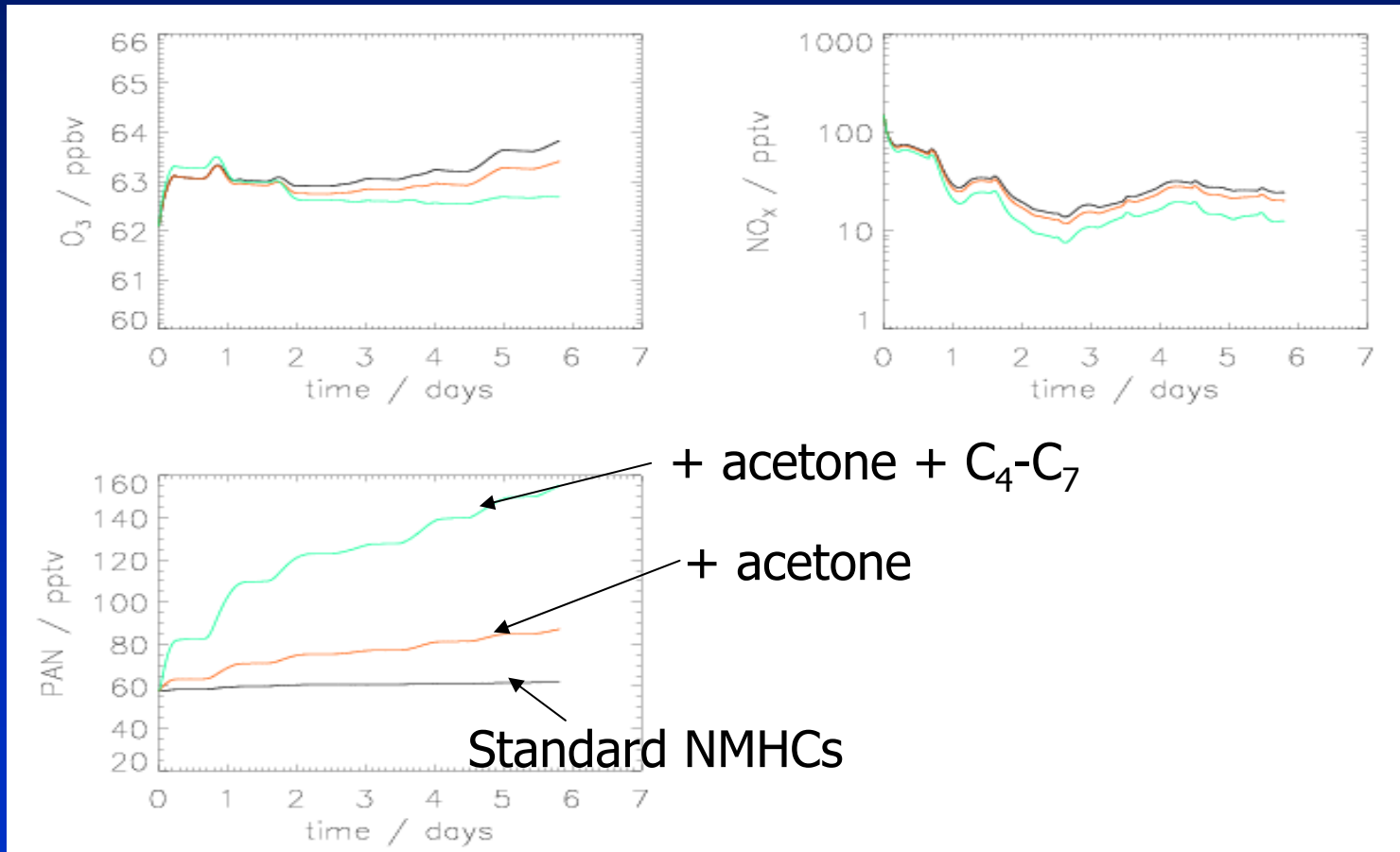


Mixing Rate



More NO_x mixed into air mass \Rightarrow *more ozone produced*

NMHC complexity



Formation of PAN through NMHC oxidation
⇒ *less NO_x available for ozone production*

Conclusions

- Trajectory forecasts are sufficiently accurate to execute intercontinental Lagrangian experiment.
- Uncertainties in observed ΔO_3 and air-mass matches influence diagnosis of trans-Atlantic photochemical ΔO_3 .

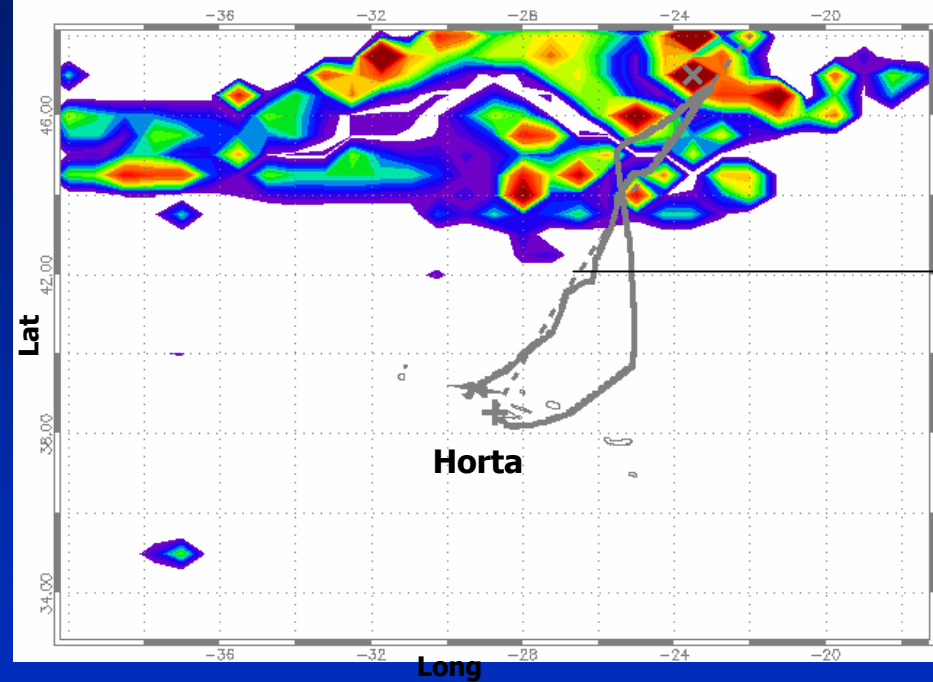
Investigations Underway

- Constraint of mixing term through hydrocarbons.
- Full Monte-Carlo analysis using Lagrangian model.
- Can reduced chemical mechanisms describe the observed change in composition or is complexity unavoidable?

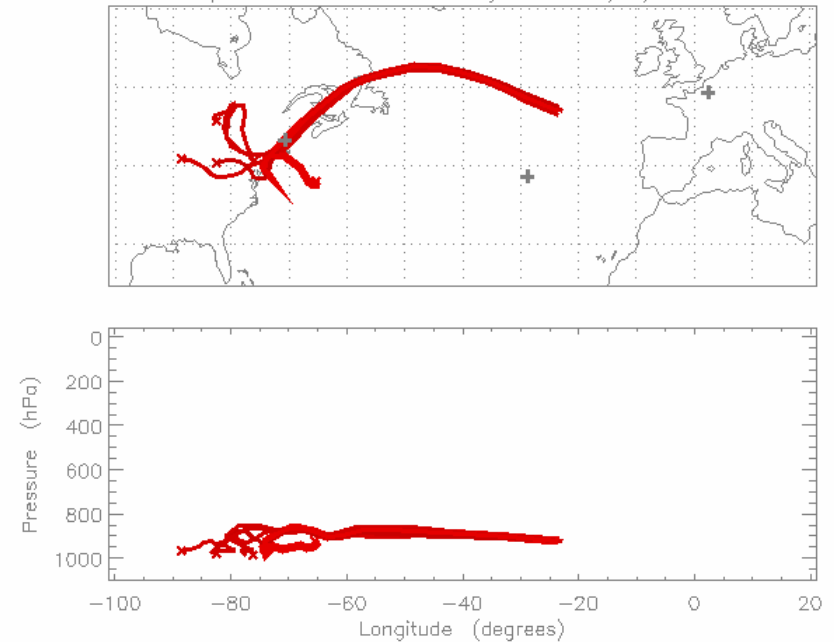
Possible multiple interception of New York polluted airmass 2

First BAE146 interception flight into NYC air 19/7

Accumulated NO_x (ppbv) 925 hPa
Release on 12UT 19/07/2004 from 000hr met forecast



Accumulated NO_x (ppbv) 12UT 19/07/2004 F+000hr
 $\lambda = -23, \phi = 47.0$ FL24, T=-7.00 days 12UT 19/07/2004 F+000hr



- NO_x emissions tracer on 925hPa on 19/7 with BAE146 (air mass relative) flight track overlaid.
- High NO_x predicted from NYC outflow.
- Obtained using RDF3D trajectories and also accumulating NO_x from EDGAR emissions inventory when in ECMWF boundary layer.

Back trajectories calculated from flight track confirm NYC as being at air mass origin

Identifying Lagrangian Opportunities

- Selected out of ~30000 trajs from each domain based on criteria:
 1. Passing within range of 2 or 3 bases (*Pease, Azores, Creil*),
 2. Accumulated NO_x emissions > threshold (*along back trajs*),
 3. Further NO_x emissions < threshold (*along forward trajs*).

(surface emissions [EDGAR] are picked up within BL as defined by ECMWF forecasts)

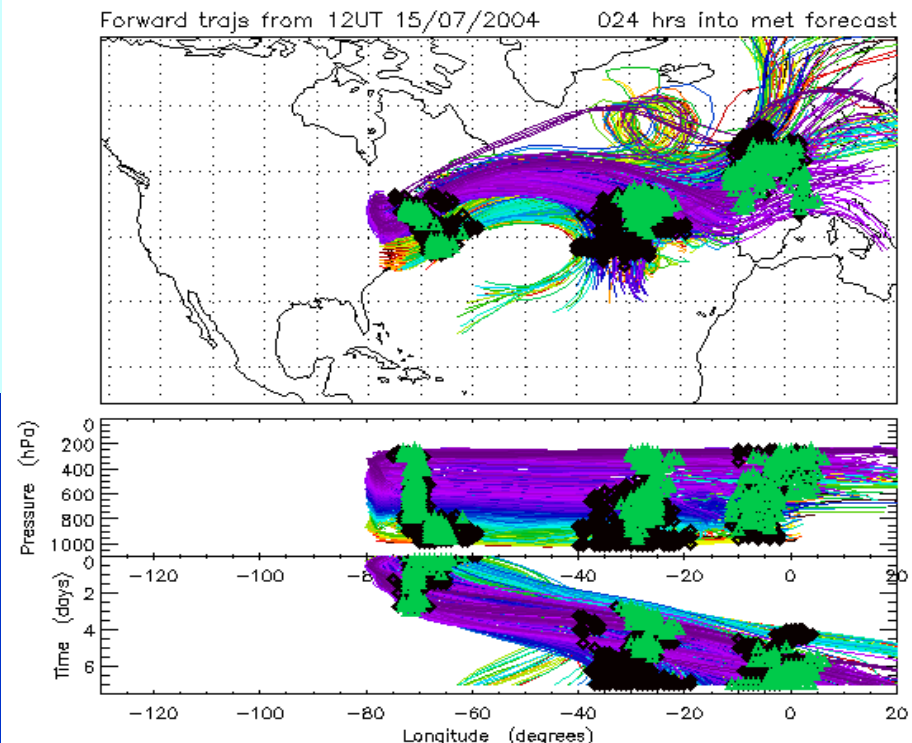
Example

Forward trajs from US domain

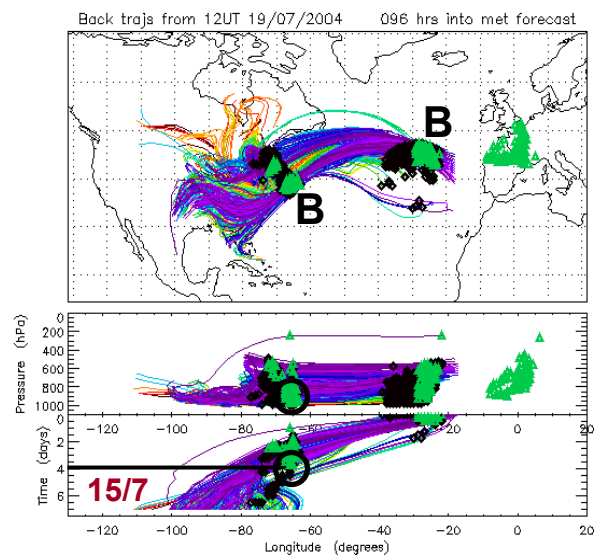
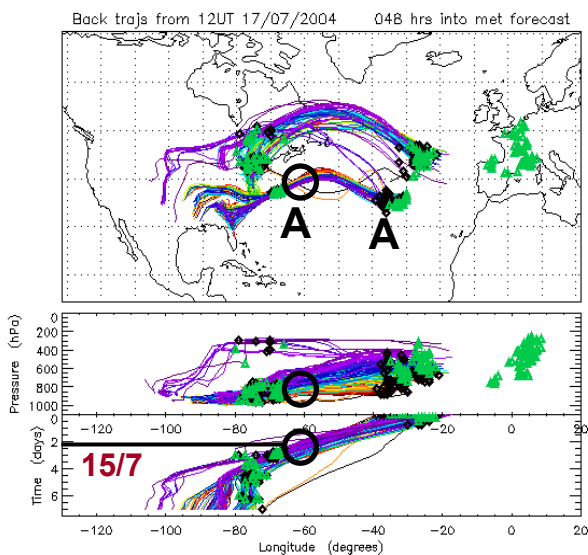
Release time 12UT 15 July 2004

Based on ECMWF forecast from
12UT 14 July 2004

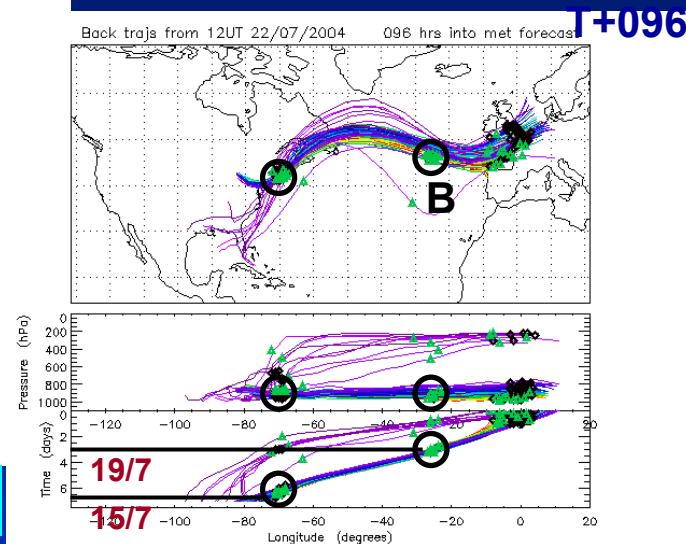
- Black diamonds – 2-point opportunities
- Green triangles – 3-point opportunities
(US – Azores – Europe)
- Red squares – 3-point opportunities
(any other order)



The first Lagrangian opportunities



Back from Europe 12UT 22/7



Back from Azores 12UT 17/7

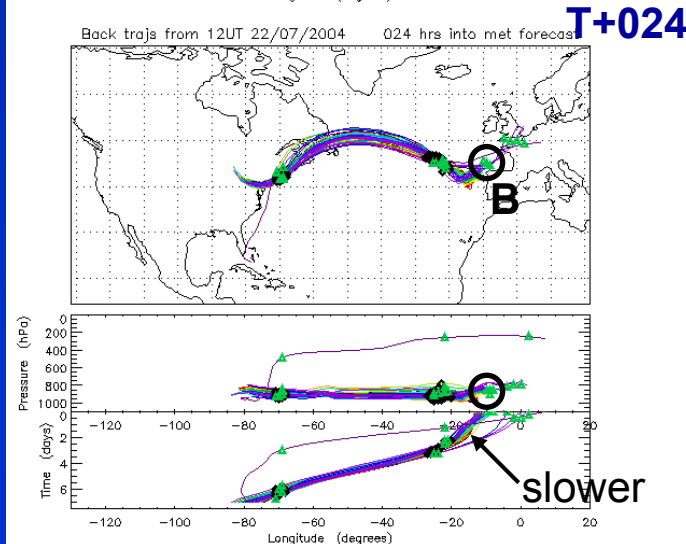
Based on ECMWF forecast from 12UT 15/7 (**T+048**)

Back from Azores 12UT 19/7

Same met forecast (**T+096**)

Note: Back trajectory calculations use wind *analyses* for dates earlier than start of ECMWF forecast.

Note similarity between trajectory forecasts with different lead times (on right). Air masses are predicted to slow down after Azores in later forecasts.



Sample collection

